Photographic Effects of Counter Discharges

Technical Report No. 1
Laboratory for Applied Biophysics
Massachusetts Institute of Technology

May, 1953

PHOTOGRAPHIC EFFECTS OF COUNTER DISCHARGES

by

K. S. Lion and G. F. Vanderschmidt

Laboratory of Applied Biophysics
Department of Biology
Massachusetts Institute of Technology
Cambridge, Massachusetts

ONR Contract N5ori-07854 May, 1953
NR 015 312

TABLE OF CONTENTS

A. INTRODUCTION, BASIC IDEA OF THE NEW PROCESS.

For many years the conventional photographic method using silver-halogen emulsions has maintained a monopoly position in experimental research and in all practical applications where a pictorial presentation of light or other actinic radiation was needed. Based entirely on empiricism and operating by virtue of impurities, the photographic process has been brought to a truly impressive level of practical performance, a level which in certain respects represents the theoretical optimum of accomplishment. Only within the last few years have we come to understand the physical phenomena involved in the photographic process. Ironically, at about the same time at which Gurney and Mott had advanced their theories of the latent image, (1938) there appeared new methods of electrical radiation detection, amplification and image formation. None of these methods is far enough developed to be a serious competitor of the photographic process, but the development undoubtedly shows the tendency to replace the photographic method by better methods for many applications.

It seems obvious to apply the methods of amplification with vacuum tubes to the problem of the detection of photons. Where the mere presence of photons is to be measured or recorded, the photomultiplier or the scintillation counter are superior to the photographic process in many respects. However, for the reception and pictorial presentation of an extended radiation pattern these methods are impractical, and other means must be employed.

This report deals with a new system for the two-dimensional reception and intensification of a radiation pattern. Its principle is the following (Fig. 1).

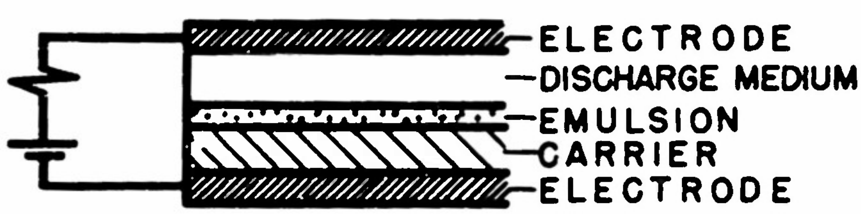


Fig. 1

10^{-3}, the gain to be expected would be of the order of $10^{-2} \times 10^{8} \times 10^{-3} = 1{,}000$. This is a rough, but probably conservative, estimate.

B. POSSIBLE PHYSICAL MECHANISMS OF INTENSIFIER ACTION.

If a photoactinic radiation strikes a photographic emulsion, part of the incident light is absorbed by the silver halide grains. The absorbed quanta lead to the formation of "sensitivity specks," and if a certain critical size of such specks is formed, the grain becomes developable, i.e., the silver halide grain can be reduced to metallic silver by developing. The number of absorbed quanta necessary to render a single grain developable varies with different emulsions, but it is in general on the order of 10. It can happen that a single quantum is sufficient to make a grain developable, while some emulsions require on the order of several hundred quanta. The number of silver atoms formed in each grain after development is on the order of 10^{10}, so that for each absorbed quantum roughly 10^{9} silver atoms are formed.

This is an enormous figure, and it may seem hopeless to try to increase the sensitivity of the photographic emulsion. However, this is not so, the emulsion can be made more sensitive in several ways:

a. It is impossible, of course, to improve the response of a single grain requiring the absorption of only one photon, since its response (namely the potential reduction of all its molecules to metallic silver by the action of one photon) already represents the theoretically optimum effect. The situation is different for emulsions in which a single grain, in order to become developable, requires the absorption of many photons. In such cases, an increase of sensitivity is possible through the above described mechanism which, when triggered by an incident photon, releases the necessary number of quanta to a grain to make it developable. The thus obtained increase of sensitivity would represent the theoretical maximum for a single grain and would not affect the resolving power, provided the discharge effect is localized to the grain.

A similar case exists for very low levels of incident radiations where most emulsions exhibit a reduced sensitivity (reciprocity law failure). This effect occurs in emulsions which require a number of quanta for the formation

Two electrodes forming a parallel plane counter are connected to a voltage source. At least one electrode is transparent to the radiation to be recorded. A discharge-sensitive material, such as a photographic emulsion on an appropriate carrier or a fluorescent substance is fastened to one electrode. If radiation enters the counter, a discharge will be initiated, which produces a dot on the photographic emulsion or the fluorescent screen many times more intense then the dot which would have been produced by the initial radiation directly.

Figure 1 illustrates only the basic principle of the method discussed in this report. A number of variations can be used. The photographic emulsion can be replaced by any other discharge-responsive material or system. The gas atmosphere in which the discharge occurs can be replaced by crystal materials. Fluorescent layers that convert the incident radiation into radiation of different wavelengths can be added, or such layers can be placed between the discharge space and the discharge responsive layer. Photoelectric active surfaces and layers can be added or photoconductive layers which change the potential distribution in such a counter when radiation strikes these layers. This study, deals primarily with the basic arrangement of Fig. 1.

Assuming that for each incident photon n electrons are produced which are able to initiate a discharge, that A particles are formed in a single discharge, and that f is the relative effect which each one of these particles formed in the discharge has upon the discharge responsive layer as compared with the effect of the incident photon, then the total gain of the amplication process is

$$G = n \cdot A \cdot f.$$

It is impossible as yet to estimate with any accuracy the numerical value of G. The value of n for the external photoelectric effect is in the order of 10^{-2}. For incident X-ray quanta converted by means of fluorescent screens into visible or ultraviolet light and these light quanta used for the production of electrons, n can have substantially greater values. The value of A is the same as the "gas ampliciation factor" customarily used in Geiger counter theory; it can attain values as high as 10^8, and for spark counters values that are even higher. The value of f has not yet been measured; assuming it tentatively as a value of

10^{-3}, the gain to be expected would be of the order of $10^{-2} \times 10^{8} \times 10^{-3} = 1{,}000$. This is a rough, but probably conservative, estimate.

B. POSSIBLE PHYSICAL MECHANISMS OF INTENSIFIER ACTION.

If a photoactinic radiation strikes a photographic emulsion, part of the incident light is absorbed by the silver halide grains. The absorbed quanta lead to the formation of "sensitivity specks," and if a certain critical size of such specks is formed, the grain becomes developable, i.e., the silver halide grain can be reduced to metallic silver by developing. The number of absorbed quanta necessary to render a single grain developable varies with different emulsions, but it is in general on the order of 10. It can happen that a single quantum is sufficient to make a grain developable, while some emulsions require on the order of several hundred quanta. The number of silver atoms formed in each grain after development is on the order of 10^{10}, so that for each absorbed quantum roughly 10^{9} silver atoms are formed.

This is an enormous figure, and it may seem hopeless to try to increase the sensitivity of the photographic emulsion. However, this is not so, the emulsion can be made more sensitive in several ways:

a. It is impossible, of course, to improve the response of a single grain requiring the absorption of only one photon, since its response (namely the potential reduction of all its molecules to metallic silver by the action of one photon) already represents the theoretically optimum effect. The situation is different for emulsions in which a single grain, in order to become developable, requires the absorption of many photons. In such cases, an increase of sensitivity is possible through the above described mechanism which, when triggered by an incident photon, releases the necessary number of quanta to a grain to make it developable. The thus obtained increase of sensitivity would represent the theoretical maximum for a single grain and would not affect the resolving power, provided the discharge effect is localized to the grain.

A similar case exists for very low levels of incident radiations where most emulsions exhibit a reduced sensitivity (reciprocity law failure). This effect occurs in emulsions which require a number of quanta for the formation of a speck. In its initial stage, before all these quanta are received, the speck is unstable and has a tendency to regress. The lower the level of the incident radiation, i.e., the longer the time between the arrival of these quanta, the greater are the chances of regression. Also in this case is it possible to increase the sensitivity by the above described discharge mechanism which furnishes all the required number of quanta at once when triggered by an incident photon. The consequence of this action would be a rise of the exposure-density (H-D) characteristic starting from the zero point, which means the elimination of a threshold level, without the sacrifice of resolving power.

b. The number of incident quanta and the number of quanta absorbed in the photographic grains are not identical. In fact, in X-ray photography, in particular with high voltage X-rays, only a small fraction of the incident radiation is absorbed in the emulsion grains (which represent an absorbing layer of a thickness of only several microns) while the greater part passes through the emulsion without contributing to the formation of a picture. In order to overcome this difficulty in X-ray photography, intensifier screens have been used and the photographic sensitivity has been increased very successfully. The effect of such an intensifier screen consists in the additional absorption of photons and their conversion, by fluorescence, into quanta of ultra-violet or visible light, which in turn act on the photographic emulsion. The limitations of such a screen are the following:

1.) The resolving power of the photographic picture is reduced. Each fluorescent point of the intensifier screen acts as a light source and produces a halo rather than a well defined spot. 2.) The energy available for the increase of sensivity is limited to the one inherent to the incident radiation. The intensifier screen makes no new source of energy available, it only uses photons which otherwise would be lost for the production of the picture. The situation is different in our method where the incident photon only triggers the discharge, while the energy is furnished by the applied voltage source. 3.) The use of the intensifier screen is necessarily limited to the range of such X-rays which are capable of penetrating the intensifier screen in front of the emulsion, or which penetrate the emulsion and the film and act on the intensifier screen in back of the emulsion. Specifically, the method is not suitable for photons in the ultra-violet-to-infrared range, while our method is applicable in this range.

c. There is a further possibility of increasing the sensitivity of an emulsion by the use of our described mechanism which, triggered by an incident photon, releases a large number of quanta. If these quanta are released to a substantial number of grains (rather than to a single grain) and make them developable, the result will be an increased optical density. The number of photons required per unit area to cause a standard optical density can be reduced considerably. The reciprocal of the number of photons required to produce a standard density is commonly used as a measure of sensitivity or "speed" of commercial emulsions and forms the basis of the Weston scale, the ASA scale or other ratings. On this basis, the "speed" of an emulsion can be increased by several orders of magnitude, but necessarily with a sacrifice in resolution.

The most suitable characteristics of a mechanism for an intensifier depends, of course, upon the intended purpose of the picture. Essentially three factors determine the usefulness of an intensifier, be it for photography, or for screen observation: the sensitivity, the resolution, and the signal-to-noise ratio.

A high sensitivity is desirable for most photographic applications, in particular for those where low levels of the incident radiation are encountered. An increase of sensitivity through effects on the single grain, as mentioned before under a, or an increase through a more complete capture of the incident photons, as mentioned under b above, is probably always desirable. Whether a sensitivity increase brought about through the increase of the number of grains affected by the incidence of a single photon, as mentioned under c, is of advantage depends upon the circumstances. Such an increase is undoubtedly useful in medical and industrial X-ray radiography, in X-ray diffraction work and in some nuclear techniques, and to a lesser degree in spectroscopy, astrophotography and some other photographic techniques.

The resolving power is limited either by the size of the photographic grain, or by the spreading of the discharge, whichever is larger. At the present time, the spreading of the discharge is probably several orders of magnitude larger than the dimension of a single photographic grain. However, it is not unlikely that the spreading of the discharge can be drastically reduced, so that the grain becomes the resolution limiting factor.

Any intensifier system decreases the signal-to-noise ratio. The problem as to the most desirable characteristics of an intensifier system to provide for the most useful intensification is very involved. The problem has been dealt with for the case of fluorescent screen intensification by Morgan and Sturm[1] and by O'Connor and Polanski[2], but may require a more thorough treatment. Our method offers an advantage over others in that it is sensitive to radiation only during the application of a voltage; its signal-to-background ratio should be higher, therefore, than the one of a system with an inherently high emulsion sensitivity.

C. PRIOR ART.

1. Electrical methods for the reception of radiation patterns.

Considerable effort has been made during recent years to increase the sensitivity of the photographic process, or to supplant the photographic method and the fluorescent screen observation by more sensitive methods.

a. One group of methods to increase the sensitivity of a fluorescent screen in X-ray fluoroscopy makes use of electronic amplifiers and electron optical processes similar to the ones used in television technique. The fundamental idea is the one of the image converter. This idea was adopted by J. W. Coltman[3,4,5,6] and operates in the following way (Fig. 2). X-rays from a

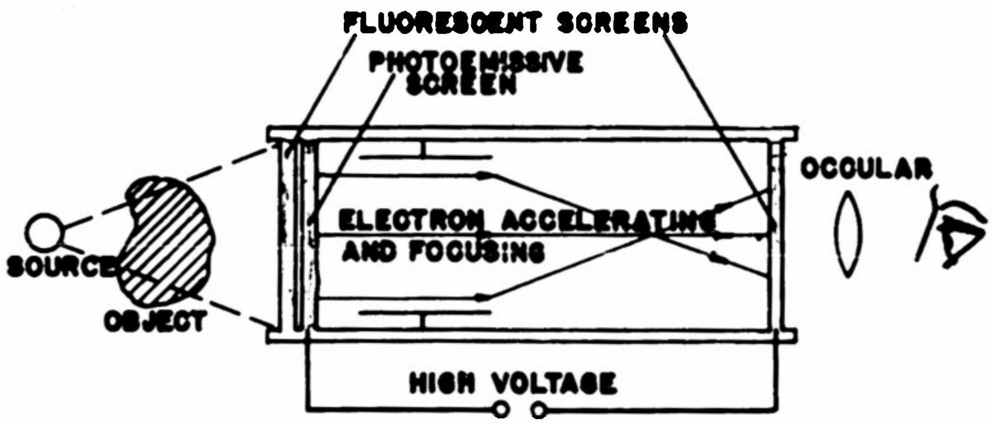


IMAGE CONVERTER

Fig. 2

source penetrate the object and strike a fluorescent screen where they are converted into light and form a fluoroscopic picture of the object. Next to the fluorescent screen is a photo-emitting surface which converts the light picture into an electron picture. The electrons are accelerated by a voltage source and are focused by an electron optical lens system. These electrons fall upon a second fluorescent screen and produce there an intensified picture of reduced size. The intensifying action is indicated to furnish a factor 20 and, since the picture is reduced to 1/5 of its size, the brightness is again increased by a factor of 25. The resulting picture is 20 x 25 = 500 times brighter than the original picture. The picture is then optically observed through a lens system and brought back to original size*

In the visible part of the spectrum a similar method has been described by R. D. Drosd, T. P. Liddard, and B. N. Singleton, Jr.[7]. The image converter is used primarily as a short-time shutter. Exposure times as short as 30 $m\mu$ sec. were obtained, and the image converter tube furnished a gain of two in intensification. Further applications of the same principle are described by F. C. Gibson.[8]

b. A different arrangement is used by Moon et al.[9,10,11,12] (Fig. 3). An electron beam from a gun scanning a target produces an X-ray beam which penetrates a system of pin holes (only one is shown in Fig. 3) in a lead shield. The scanning X-ray penetrates in succession different parts of the object and then falls upon a fluorescent crystal. The light output from this crystal is viewed by a photomultiplier, and the output from this photomultiplier is amplified and connected to the intensifier grid of a kinescope. The electron beam in the kinescope is synchronized with the electron beam in the X-ray tube, and the image is thus synthesized on the screen of the kinescope.

The last publication on this system describes it as still in an early experimental stage, but having produced promising results on a small scale. The

*Comparison with other methods should be made on the basis of the same picture size. An optical reduction in size with the accompanying result of an increase in brightness is possible, of course, to any desired degree in ordinary fluoroscopy.

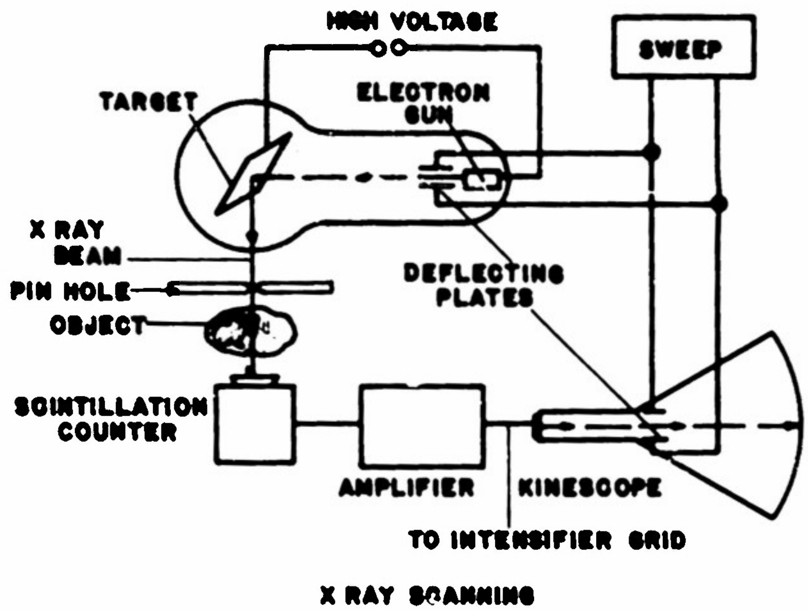


Fig. 3

method has been criticized by Morgan[13] on the grounds that while the total amount of X-rays produced within the X-ray tube is large (100 KV at 100 mA), the amount of radiation penetrating through the pinholes in the lead shield, passing through the body of the object and finally reaching the fluorescent screen, is in the order of only 10^3 photons per square millimeter per second. That is a very small amount, and the signal-to-background figure is therefore small. This fact may considerably reduce the usefulness of Moon's method.

c. A third group of electrical methods to overcome the limited sensitivity of the fluorescent screen or of the photographic method makes use of electronic and nuclear counters. The application of counters for the detection of small X-ray intensities has been well known since the discovery of the counter in 1909. Through scanning methods (usually mechanical scanning) it is possible to obtain the intensity distribution in an X-ray beam. The application of multiple counters for quasi-optical purposes is a well-known technique in the field of cosmic radiation where several of such counters are frequently lined up and used with coincidence methods to determine the angle of incidence of incoming radiation.

A method using multiple counters and a mechanical scanning system for the detection of two-dimensional radiation patterns has been described recently by W. V. Mayneord and E. H. Belcher.[14,15] This "method of making visible the distribution of activity in a source of ionizing radiation" operates in the following way: A well-canalized counter receives radiation from a small angular region of a radioactive body. Two motor-driven cams give the counter horizontal and vertical movements, enabling it to scan the radioactive body. The instantaneous counting rate will be proportional to the radioactivity "seen" at any moment. The radioactive picture received in the scanning counter is passed through a pulse shaping circuit and applied to cathode ray tube indicators. Rotary switches, attached to the driving motor of the counter, apply steps of voltage to the cathode ray tube deflecting plates appropriate to the position of the scanning head, thus producing a matrix for reception of the radioactive picture." (Author's abstract.) A reproduction of some results obtained with this method is shown, together with the schematic diagram of the method, in Fig. 4, page 56. The sensitivity of the method is probably high, but the mechanical and electrical requirements are considerable. The resolving power as well as the signal-to-noise figure will probably be much lower than the one obtained in our method.

A somewhat similar method has been proposed by F. E. Haworth.[16] It consists of a great number (4,400) of Geiger point counters, every one of them having an individual "point" (usually anode) while the cathode is common to all these counters. Every point is connected to a separate resistor and a separate three-electrode glow lamp indicator, and all points are connected through each glow lamp, to a common voltage supply. Figure 5 shows the schematic diagram. When ionizing radiation falls into the critical volume in front of any one point, a counter discharge occurs, and the glow lamp connected to the respective counter lights up. No results obtained with this method are indicated, and it seems to be somewhat doubtful whether the method lends itself readily to practical applications. 4,400 picture points are perhaps sufficient for orienting quartz crystals, but for pictorial presentation at even modest resolution, a much larger number of picture points would be required, which would necessitate a correspondingly large number of Geiger points, of resistors, of glow lamps, and of connecting lines. However, the proposal is interesting and shows

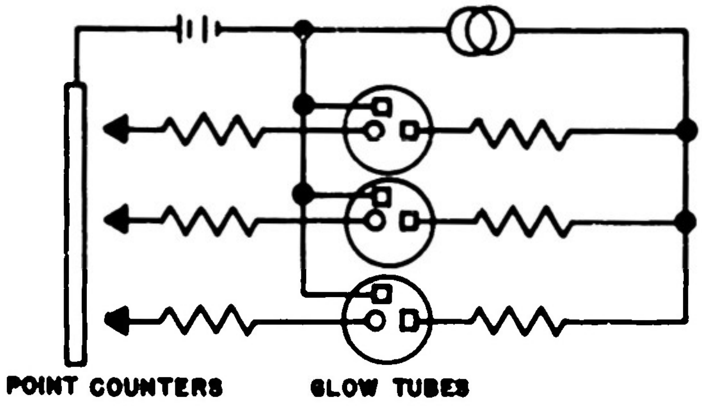
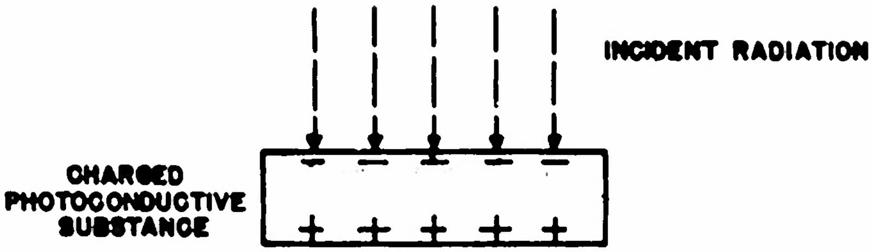


Fig. 5

to what extent technical complications are tolerated in order to solve the problem of pictorial reception at high sensitivity levels

d. A further group of electrical methods for photography aims perhaps less at an increase of sensitivity over the photographic process using silver halogen compounds, but rather at an elimination of the wet chemical process of photographic development. The method is apparently based on an early experiment by Zeleny,[17] who succeeded in making the trace of an electron beam sweeping over the face of a cathode ray tube visible by dusting a dry powder on the external surface of the screen. The powder adhered to the part of the face which was charged by the electron beam, and the trace of the beam thus became visible. A somewhat similar method has been developed for photographic processes by C. F. Carlson,[18] and is described under the title "Electrophotography" by N. Langer.[19]

The application of this method in the field of X-ray detection (xeroradiography) has led to interesting results and is described by R. C. McMaster and R. M. Schaffert[20] and in more detail by McMaster[21] (Fig. 6). A layer of a photoconductive substance (e.g., a selenium compound) is electrically charged. In complete darkness such a layer has a high resistance and electrical charges on it may be kept for several hours without losses through leakage. When exposed to radiation, the carrier becomes locally conductive and a discharge occurs at such parts where the rays strike the plate. The rate of loss of charge is proportional to the local intensity of the X-rays. The distribution of the

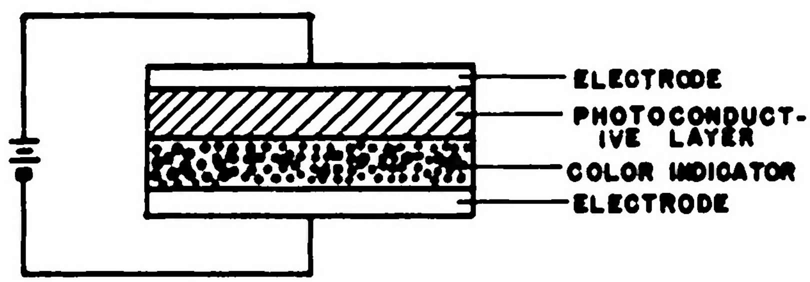


XERORADIOGRAPHY

Fig. 6

radiation given by the remaining electrostatic image is made visible by a pigmented powder which adheres to the charged parts of the film. The image may be viewed within a few seconds after X-ray radiation, it may be temporarily fixed, or it may be transferred to an adhesive coated paper. The publication by McMaster indicates that the speed obtained with this method is about 2 to 7 times that of a type M radiographic photo material. This is a considerable accomplishment, but it is questionable whether a comparison with a low sensitivity fine grain emulsion is justified. The resolving power is less than that of a fine grain silver halogen emulsion, but it is satisfactory for a great number of practical applications.

In this same group belongs an electric-photographic process proposed by F. Goldmann[22] and operating in the following fashion (Fig. 7): A photoconductive layer and a chemically prepared layer containing a color indicator are placed between two electrodes. The two electrodes are connected to a voltage source. Incident radiation causes an increase of conductivity of the photoconductive material. The latter material is such that when current passes through



GOLDMAN METHOD

Fig. 7

it, it undergoes a chemical reaction with the adjacent layer, and the color indicator makes this reaction visible. There seems to be no indication in the literature that the Goldmann method has ever been used. The sensitivity, resolving power or other physical properties of this arrangement are not given either, but it is unlikely that the sensitivity of this method can ever approach the one of a silver bromide emulsion.

e. Of an entirely different character (and included here only for the sake of completeness) are the methods of ordinary photography of sparks produced in counters. One of these methods has been introduced into the literature by Greinacher[23] in connection with his work on spark counters. The spark counter and an arrangement for the photographic recording of sparks is shown in Fig. 8.

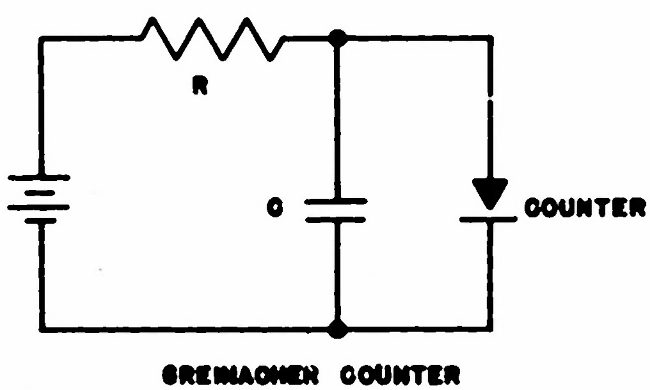


Fig. 8

The spark counter differs from the ordinary Geiger point counter by the presence of a capacitor (order of .001 MMF) in parallel to the discharge gap. The counter is not of the self-quenching type, but requires, like a resistance quenched counter, that the voltage across the counter be diminished below the minimum sustaining voltage. This requires a discharge of the capacitor, a process which is connected with the flow of a considerable amount of electricity. The discharge has the character of a powerful spark which can be heard directly or recorded photographically. Of course such a record does not furnish a two-dimensional picture, but only a series of dots on a moving film.

Different from this method is one developed by T. H. Johnson.[24] His method operates in this way: A foil of indium which has been activated by slow neutron irradiation is mounted on the cathode of a plane parallel counter and is covered with a thin foil of pure tin. A piece of conductive glass (NESA-glass) acts as anode. A discharge of this counter is characterized by a tiny spark and a brilliant glow on the cathode. This light phenomenon is observed and photographed through the glass anode, thus outlining the pattern of activation on the indium.

2. Electrical effects on photographic and fluorescent materials.

After many years of experimentation, today it seems a matter of course that counter discharges produce traces on photographic emulsions. However, the literature on this subject is sparse in general, and with respect to counter discharges and their effects on photographic emulsions, no references seem to exist.

a. Probably the most widely investigated effect of electrical discharges on photographic plates or films is the phenomenon of "Lichtenberg's Figures" first observed on Daguerre-type photographic plates in 1851. The arrangement for producing such discharges is shown in Fig. 9. The voltages are in the order of several kilovolts. Depending upon the polarity of the electrodes, different characteristic figures arise on the photographic emulsion, which according to a comprehensive study of the Lichtenberg Figures by F. H. Merrill and A. von Hippel[25] represent a photographic record of the light emission of the discharge. A Lichtenberg Figure is shown in Fig. 10, page 56. The Lichtenberg Figures have found some practical application in lightning research and measurement of high voltages (Klydonograph, Keinath[26]).

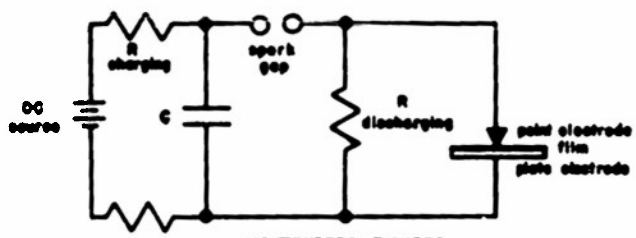
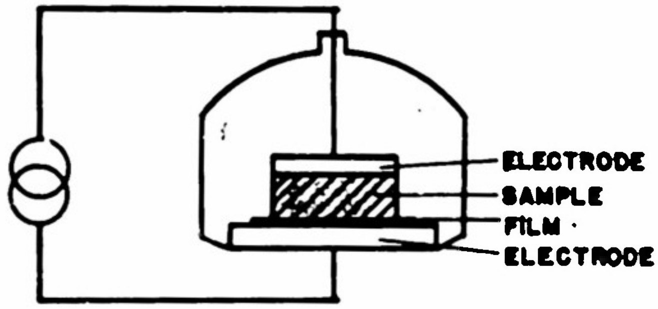


Fig. 9

b. An entirely different kind of an effect of discharges on photographic materials has found some notoriety, also under the name "Electrophotography." The effect, first observed by Gemant[27] in 1931, comes about in the following way (Fig. 11): A high voltage source (AC, 60 cycles, order of magnitude 50 kV) is applied to two electrodes, one of which is grounded. Adjacent to the grounded



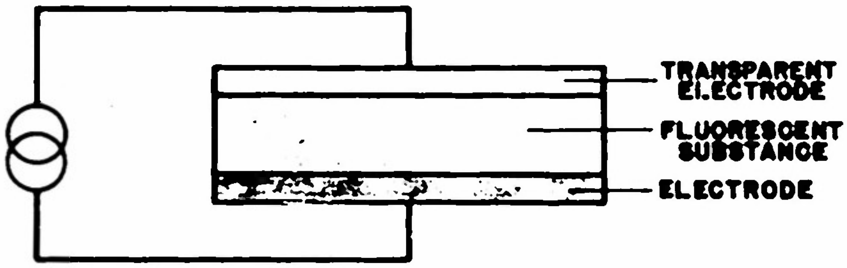
GEMANT ELECTROPHOTOGRAPHY
Fig. 11

electrode is a photographic plate or film, and next to it a piece of an insulator and the high voltage electrode. The entire arrangement is shielded from room light and from electrostatic stray fields. When the voltage is raised, discharges occur at places where the insulating material is defective. It is thus possible to examine insulating materials photographically. This method has been used repeatedly and is described furthermore by Gemant,[28] Thomas,[29] Akahira and Kamazawa,[30] and lately by Mason.[31] Prat and Schlemmer[32] have used it to examine surface properties of biological materials.

In connection with our own work, we have examined the Gemant method in order to ascertain whether – with the insulating layer replaced by appropriate materials – it would be suitable for the purpose of radiation detection. We also have used the method to examine insulating materials for application in counters. Figure 12, page 56, shows an example of exposures with mica obtained with this method.

A variation of these methods is described by Zahradnicek[33] in a paper entitled "Photochemical effects produced by Maxwell's current." It differs from the one of Gemant primarily by the application of higher frequencies (10^2 - 10^6 cycles per second). The voltages used are of the order of 10^4 volts, the current thus produced is "several milliampere" (displacement current?). The author states that "the kinetic energy of ions or electrons in the current is changed into photochemical energy on the photographic plate, as long as the energy of an electrode particle is greater than $2 \cdot 10^{-12}$ erg, i.e., 1 eV." That is a very general statement and does not express anything about the mechanism involved. With a voltage of 10^4 volts applied, a gas discharge process is likely to occur.

c. Similar to the last group as far as the physical setup is concerned, but different from it in the physical mechanism involved is a process called "Electroluminescence." The effect was observed first by Destriau[34] and can be demonstrated with a setup as shown in Fig. 13. A luminescent substance,



ELECTROLUMINESCENCE

Fig. 13

such as a conventional phosphor of the zinc sulphide type, is placed between two electrodes of which at least one should be transparent. If an alternating voltage is applied to the electrodes, an excitation of luminescence under the influence of the applied electric field can be observed. Fields used are in the order of 10^4 to 10^5 Volts/cm. A transient effect can be noted when DC fields are applied; with AC fields the effect increases with increased frequency and increased fieldstrength. The light emission is of periodic nature, having twice the frequency of the applied voltage, and a time delay between the peak of the applied voltage and of the emitted light. In contrast to the methods mentioned under a and b above, the effect does not seem to be due to a gas discharge produced by the applied field, but rather to an effect of the field upon electrons trapped in impurity centers of the phosphor. It is interesting to note that apparently a similar (or opposite) effect of an electric field upon photographic emulsions has not been observed or described in the literature, although one should assume that an effect of an applied electric field upon an exposed silver halide crystal might exist. The process of electroluminescence has recently found some application for illumination purposes.[35,36,37]

d. In this collection of references on the effects of electrical discharges and currents on photographic and luminescent materials, belongs also an account of the impact effect of electrons and ions upon layers of such materials. A large amount of information is available on this topic because of its practical importance in the fields of television, radar, cathode ray tube and mass spectrograph techniques. However the majority of the work done in this field has little connection with the present problem, primarily because most experiments are made with electron and ion beams of high velocity, which most certainly are not present in counter discharges.

As far as the photographic effect of electrons is concerned, one should assume from energy considerations that any electron with an energy greater than one e Volt is capable of producing a photochemical effect in a silver halide grain. Greater velocity would only enable the electron to penetrate to a greater depth of the emulsion. Thus, it is understandable that for the investigation of slow electrons, mostly photographic emulsions of low gelatine content have been used. It has been stated (Klemperer[38]) that the photographic effect of electrons comes about in two different ways: first, through the direct action of the electron upon the silver bromide crystal, and second, through an indirect effect via a radiation produced by the electron at the surface of the emulsion. The latter process is of particular significance for the photographic effect of very slow electrons. For instance, it is customary to sensitize photographic emulsions for slow electrons by applying fluorescent materials on their surface.

Considering the variety of physical mechanisms involved, it is understandable that the relationship between the final density of the photographic emulsion and the number and velocity of the incident electrons is rather complicated, in particular at low velocities. However, certain trends are fairly well established:[39,40]

1. The density produced by fast electrons of several thousand e Volts is strictly proportional to the product (i• t) (in Coulombs). At low electron

velocities the density is proportional to $i \cdot t^p$, where p is in the order of 0.9.

2. The H-D curve for fast electrons shows no threshold value and no toe part, the density increases from zero, first steeply, and approaches a saturation value. At such high velocities of several kilovolts the relationship between the photographic density and the incident current as well as the voltage applied for the acceleration of the electrons seems to be well established and is described, apparently with satisfactory accuracy, by the Silberstein[41] equation. The curve for slow electrons shows first a slow increase in density due to the direct effect of the electrons on the AgBr on the surface of the emulsion, later a steeper increase resulting from secondary effects of these electrons which affect the deeper layers of the emulsion.

3. The influence of the velocity of the electrons upon the density follows a very complicated manner. It has been claimed[42] that electrons of an energy less than 22 e Volts do not produce any photographic effect, but that the effect increases steeply above 35 e Volts. From 60 to 200 volts the sensitivity of the photographic emulsion is independent of the velocity of the incident electrons. The picture is further complicated at higher electron velocity, but this range is of no interest in connection with the problem treated in this report.

The situation with respect to the photographic effect of positive or negative ions is even more complicated than the one of electrons. References in this field are mostly found in connection with literature on Canal-rays. In general it has been found that:

1. The photographic effect is proportional to the number of incident particles, but seems to be independent of the charge, at least at velocities down to several hundred volts.

2. The sensitivity for light of a particular emulsion gives no indication as to the sensitivity for ions. Schumann plates have been used successfully for the detection of slow ions.

3. Particularly high sensitivities have been found for hydrogen and potassium ions (Bainbridge[43]).

Since an ion presents a certain amount of potential energy of a magnitude of at least the ionization potential, one should assume that such an ion carries the necessary energy for a photochemical effect, even it it strikes the silver bromide with thermal velocity, and much more so if its velocity is in the order of several volts.

The effect of luminescence in phosphors produced by the impact of electrons is called cathodoluminescence. The process has a rather low efficiency ($\sim$10% of the energy of the incident electrons are converted into light output as compared to 95% for light excitation of phosphors). The luminescence intensity per unit area of a phosphor screen is proportional to the current density at low incident current densities, and shows a saturation effect at higher current densities. The luminous emission increases exponentially with the applied voltage. However, some phosphors seem to have a lower limit, a voltage level below which no light emission can be noted (dead voltage). Transient phenomena can complicate the physical process so that different modes of operation exist for DC and for pulse excitation.[44]

D. EXPERIMENTS, GENERAL CONSIDERATIONS

The simplicity of the physical arrangement and the operation of our radiation detection method is in sharp contrast with the complicated internal mechanism involved. H. Friedman[45] describes the Geiger counter mechanism very adequately as "deceptively simple." The puzzling variety of the phenomena observed, such as proportional pulses, Geiger pulses, sparks, discharge forms that are triggerable and others that are self-sustaining, such that produce marked photographic effects, and others that do not; the dependence of these discharge forms on seemingly unimportant variations of the experimental set-up or on traces of impurities, all these observations have been complicated in our initial experiments. The situation has been worsened by the enormous amount of existing literature on the subject of gas discharges and counters, with different schools using different nomenclatures so that a comparison between them or a correlation to our own work is difficult or altogether impossible. Some of the older papers in the field of gas discharges and counters, in particular in the field of point counters, are erroneous and misleading, and only slowly have we come to discriminate (as much as is possible) as to what is acceptable and what is not. Literature in the field of plane parallel counters is very sparse, and on counters with semiconductors (photographic film) it is almost completely absent.

Under these circumstances it may seem best to break down the entire phenomenon of image formation in a counter into the single processes involved, and to investigate the influence of each independent variable upon a "unit process." The disadvantage of this approach may be the relatively large number of experiments required; involved in the operation of a counter for photographic image formation is the effect of about twenty independent variables (see Table I) determining the character of the counter action, which in itself is described by seven physical magnitudes. However, the situation is not as discouraging as the preceding may imply, and it will be necessary only to establish the basic trend of the counter operation by experiment in order to find the connecting links with the existing literature, and to apply theoretical methods.

Some of these experiments are described in the following. As stated before, the greatest difficulty in all this work is the establishment of reproducible experimental conditions. At times this has been exceedingly time consuming, and conditions which were adequate to produce satisfactory pictures were grossly inadequate for measurements, while at other times, when working under such clean conditions that satisfactory reproducible measurements could be obtained, the picture vanished almost completely. Of the great amount of material obtained in these experiments only some representative examples are included in this report.

TABLE I

Independent variables

- Applied voltage
- Polarity
- Electrode distance
- Gas, type
- Gas pressure
- Quenching agent
- Concentration of quenching agent
- Shape of electrodes
- Mat. of anode
- Mat. of cathode
- El. resistance in series
- Parallel capacitor
- Type of incident radiation
- Intensity of incident radiation

Dependent variables

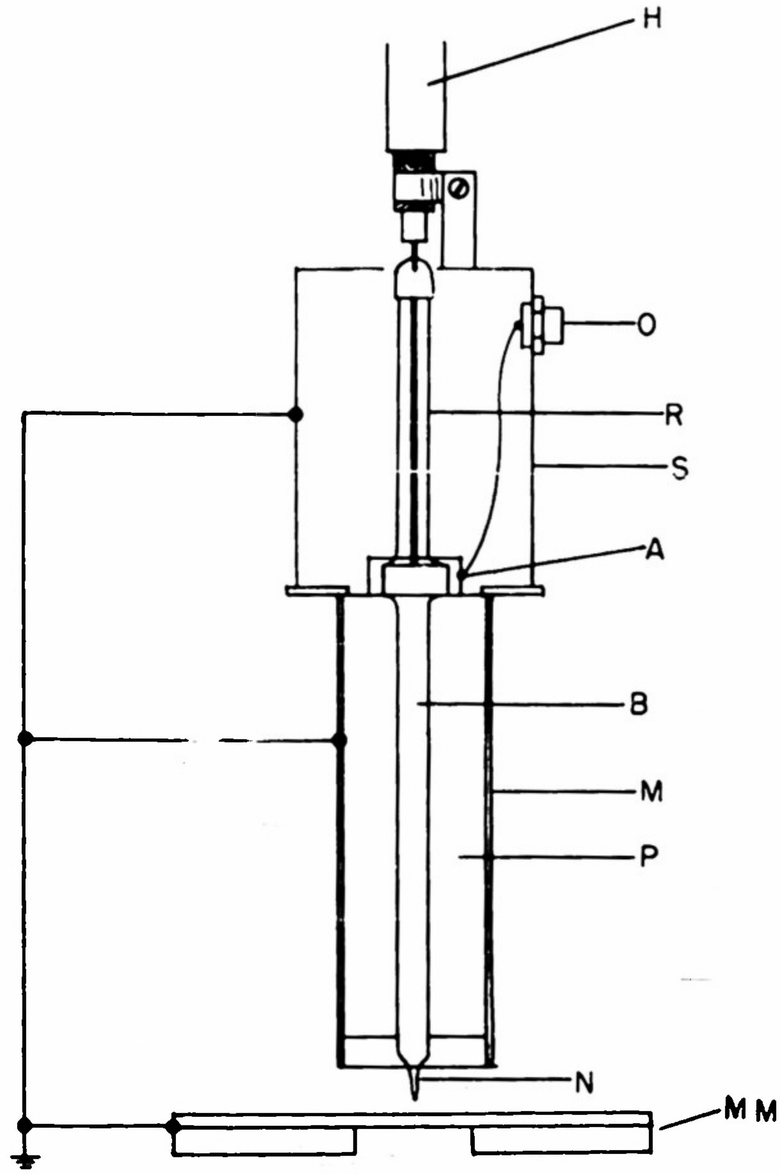
- Counting rate
- Pulse size
- Triggerability (background)
- Photographic effect (blackening)
- Photographic effect, resolution
- Recovery time
- Quantum yield

1. Point counters

As far as the experimental setup is concerned, it would be best if in these experiments a parallel plane counter could be employed and triggered at one point only so that the single discharge process could be observed, or the accumulated effect of multiple discharges occurring at the same point. Unfortunately this implies considerable experimental difficulty, not only because of the finite cross section of the collimated X-ray beam, but also because of the large electrical resistance of the presently used commercial photographic material. This has the effect that a discharge arising at one point neutralizes the surface charge so that the next occurring discharge is not likely to occur at the same place. To some extent the difficulties can be overcome in point-to-plane counters, where the discharge is forced to reoccur at the same point or, at least, within a very small area. Most of the experiments, therefore, were carried out in point-to-plane counters. Such a counter offers the additional advantage that any visible discharges can be readily observed, and that no adjustment for parallelism of the plates is needed. Its disadvantage is the fact that because of the different geometry the discharge character is different from the one in a parallel plane counter, so that the results from these experiments need careful interpretation if they are to be applied to the parallel plane counter.

a. Discharge characteristics, general behavior of the point counter

The first experiments on point counters were made in air without quenching agent. The presence of large amounts of electronegative gases in air (O_2, H_2O) along with its varying constituency makes air a poor choice for gas discharge experiments. However, the initial experiments gave a valuable indication of what to expect in a general way. The apparatus used in studying the effects is represented in Fig. 14. For the experiments with pure gases (argon and nitrogen) and quenching agents a modified version of the counter was used, as shown in Fig. 15, page 58. In this arrangement the needle is surrounded by a bell jar through which a stream of gas may be passed. The

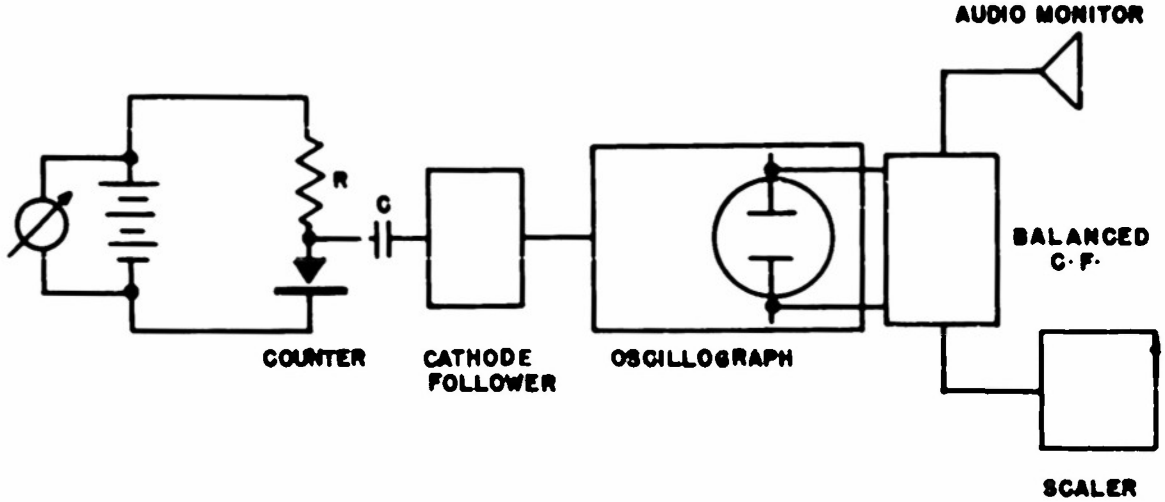


H	high voltage
O	output
R	resistance
S	shield
A	annular ring coupling condenser ($6\mu\mu F$)
B	brass rod
M	microscope tube
P	plastic insulator
N	needle
MM	microscope stand

Fig. 14

counters were used in the experimental setup indicated in Fig. 16. Any variation in voltage across the counter gap is observable on the oscilloscope and may give rise to a count on the scaler. The cathode follower is connected between the counter and the oscilloscope as an impedance changer which helps to preserve the sharp rise time of any pulses which may appear.

Three distinct types of discharge were noted without the presence of photographic material on the cathode. At any setting of distance from point to



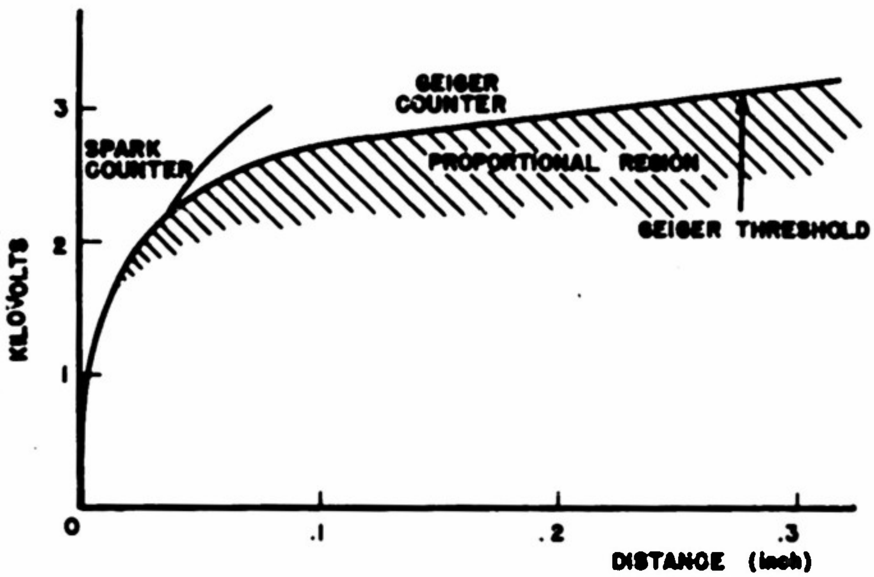
EXPERIMENTAL SET-UP

Fig. 16

plane, the first voltage variations to be observed as the supply voltage is raised are small sharp pulses. They appear when a radioactive source is held near the counter gap; if the source is removed, the pulses disappear. These pulses appear at a poorly defined threshold voltage, their average size increasing rapidly from zero as the supply voltage is increased. They are of different sizes, have a sharp rise time (less than a microsecond) and a decay time dependent on the coupling circuits. (See Fig. 17a, page 56.) The largest average size is about ten millivolts. These pulses are termed proportional pulses.

A second type of pulse appears at a slightly higher voltage than the ill-defined threshold for proportional pulses, provided the needle to plane distance is about ten times the needle point diameter or greater. This pulse is about one hundred times larger than the proportional type pulse (one volt) and has a rapid rise time. The pulses are all identical in size. (See Fig. 17b, page 56.) As with the proportional types of pulse, these pulses appear only in the presence of ionizing radiation. The pulses have a finite value at the threshold and increase in size only slowly with increasing supply voltage. The counting rate due to external radiation goes up very rapidly with increasing supply voltage beyond the threshold however, and at some point the discharge becomes self-sustaining, i.e. maintains itself independently of external radiation. This type of pulse is identified as the Geiger pulse.

The third type of discharge is a spark. The spark occurs at a slightly higher voltage than the proportional pulse threshold for needle distances less than about 10 times the needle diameter. It also occurs at greater distances when the voltage has been raised above that necessary for a self-sustaining Geiger discharge. The voltage pulses produced are identical to the Geiger pulses in shape, but much larger, approximately 50 volts. Very close to their threshold the sparks are triggered by external radiation, but if the voltage is increased slightly beyond this, they form a self-sustaining discharge. The three regions are indicated in Fig. 18 for the case of air as a gas and for atmospheric pressure.

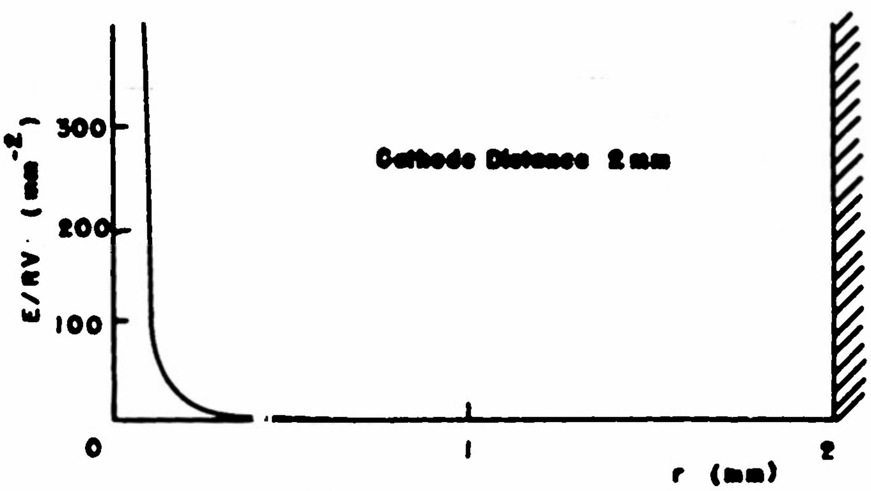


POINT COUNTER IN AIR

Fig. 18

The explanation for the described behavior is closely analogous to the one in an ordinary Geiger counter of cylindrical geometry. The space between the needle point and the plane is occupied by a gas consisting of neutral molecules. If an electric field exists here, there will be no effect. If, by means of external radiation, a gas molecule becomes ionized, then particles carrying an over-all electrical charge exist in the gap, and current will begin to flow in the circuit; the current being simply the motion of the induced charges. The presence of current in the circuit as indicated by a drop in the voltage across the counter is then direct evidence of ionization in the gap.

The presence of the electric field introduces the possibility of cumulative ionization, once an initial ionizing event has occurred. The immediate products of an ionization are usually a positive ion and one or two electrons. The positive ion will drift towards the cathode, and the electrons towards the anode. The positive ions and electrons will be accelerated by the field in opposite directions, and will experience elastic collisions. Without going too deeply into their exact motion, it seems that the positive ions will drift much more slowly than the electrons due to the enormous difference in mass. In the case of a positive point, the electrons are attracted in a direction of rapidly increasing field strength. If for instance we assume that the needle point is effectively a sphere, for the case that the needle radius is less than 1/10 the distance from point to plane, the field distribution is practically $1/r^2$, as seen in Fig. 19.



Field Distribution Around A Point

Fig. 19

As the electrons enter the region of more intense field strength, they will pick up sufficient energy to make ionizing collisions. If the field strength is sufficiently high, the electrons may achieve enough energy to ionize at each collision. If α is the number of secondary ion pairs produced by each centimeter traveled by an initial electron then the number of ion pairs formed by N_o initial ion pairs is

$$N = N_o e^{\int_{R_2}^{R_1} \alpha \, dr} \tag{1}$$

where R_2 = radius of the surface of the needle point, R_1 = radius at which the initial ion pairs are formed. Since α is a function of the field strength it is also a function of r, and we must use the integral form of this equation.

We can identify the first sort of voltage pulse observed with a discharge of this sort, in which an initially formed electron produces an avalanche of ionization. The voltage pulses appearing across the counter will be in some way proportional in size to the amount of ionization in the gap. We expect a poorly defined threshold where just enough cumulative ionization is being produced so that the pulses are large enough to appear over the noise in the system. We expect the pulses to be of different sizes, since N is directly proportional to N_o, the amount of initial ionization. We expect the average pulse size to increase rapidly with increase of the counter voltage, due to the exponential form of equation (1). And of course, if there is no initial ionization, there will be no discharges, thus the pulses cease when the external source of radiation is removed.

In the case of the proportional pulse, once all the charge is collected by the electrodes, the discharge ceases, as there is no fresh ionization to sustain it. There is a possibility, however, that the discharge may produce a fresh supply of electrons by some secondary process, and will become self-sustaining. (These new electrons may be liberated from the cathode by photons generated in the discharge[46], or they may be generated in the gas itself by photoionization.) At some critical value of field strength the proportional pulse avalances will become sufficiently intense to cause new electrons to be formed at the cathode or in the gas gap, and the discharge can continue, building up in size. Eventually the building up of the slow moving positive ions in the gap will reduce the field strength around the needle point to a value below which it is not possible for the secondary process to remain active, and the discharge will cease. Thus the amount of ionization produced in such a discharge, along with the voltage pulse which it will induce, is independent of the amount of initiating ionization, and for any setting of counter voltage we can expect all the pulses observed to be the same size and considerably larger than the proportional type pulses. Further, we can expect the pulses to have a sharply marked threshold due to their finite size. This describes the second type of pulse observed, and it is given the name self-sustaining or Geiger discharge. Finally, if the field strength is increased beyond a certain point, a great many positive ions will have to accumulate before the discharge will cease, and there may be enough so that in striking the cathode when they are eventually collected, an electron may be released, causing the process to reestablish itself, and leading to the continuous discharge.

The spark is not of overwhelming interest, for the time being we might think of it simply as the form which the Geiger pulse takes when the field strength across the whole gap is sufficiently high to sustain ionizing collisions at every point in the gap.

b. "Counting rate vs. voltage" curves.

If the counter is adjusted to any fixed distance and the voltage is raised, no counts will be observed below a certain level. At the Geiger threshold a number of counts will be observed and the counting rate increases very rapidly with increased voltage (A in Fig. 20). Then follows a region (B), of stable counting in which the counting rate increases less rapidly with voltage. This region is characterized by a constant slope when plotted on semi-logarithmic paper. It follows then a very sharp increase in counting rate (C) at some voltage, marking the point at which the counting rate becomes self-sustaining, i.e. untriggerable. Further increase of the voltage generally leads to bursts of discharges, occational sparks, and finally, continuous spark discharges.

We call the region of stable counting the plateau, even though it is not a plateau as observed in cylindrical Geiger counters. Fig. 20 shows such a characteristic curve for a mixture of argon and methyl alcohol.

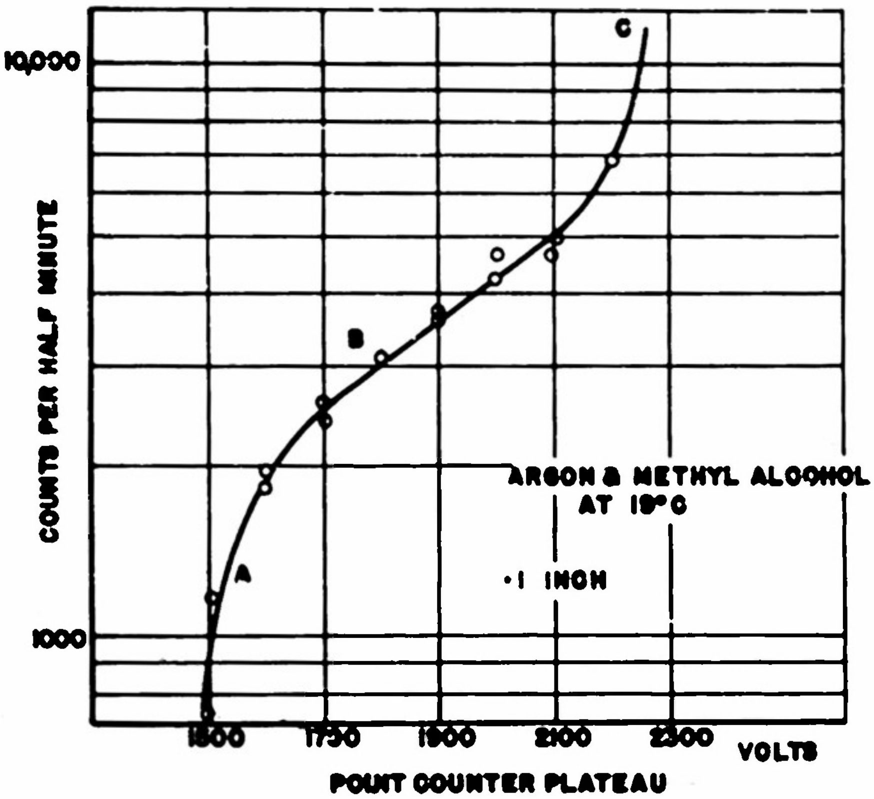


Fig. 20

c. Influence of the electrode distance:

As the distance between the needle and the plane is increased, two changes take place. First, the entire curve is shifted to the right, i.e. towards higher voltages. This, of course, is to be expected, since a separation of the electrodes reduces the fieldstrength, and higher voltages have to be applied to produce those fieldstrengths at which counting takes place. Second, the center of the plateau is shifted upwards (as well as to the right), i.e. the counting rate at a given voltage above the threshold is higher for counters operated at larger distances. This may be interpreted as a change in the shape of the field distribution with larger distances which results in an effective increase of the critical counting volume of the counter. Third, the plateau part is more extended at larger distances. In fact, at very small distances the plateau disappears completely, and the number of counts increases very rapidly with the applied voltage throughout the range of Geiger discharges. The disappearance of the

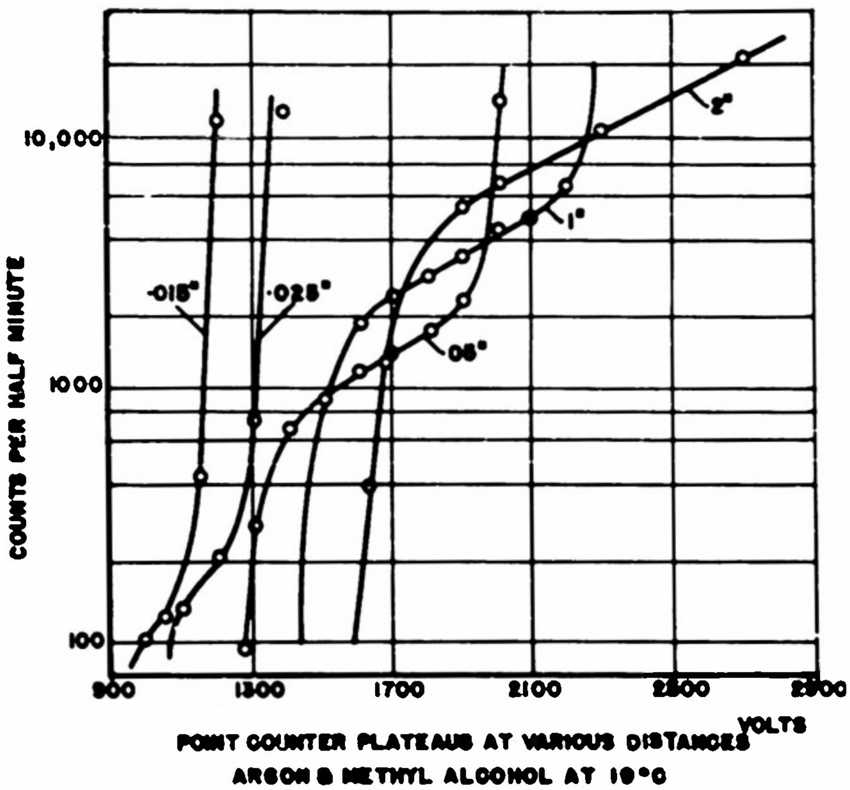


Fig. 21

plateau at small distances may have its cause in the fact that the path between point of origin of photons in the discharge (near the needle point) and the place where these photons produce secondary electrons (the cathode) has become so small, that satisfactory absorption of these photons is no longer possible. It will be seen later on that this effect, namely the disappearance of the plateau at small electrode distances can be compensated partly by increasing the percentage of the quenching action, or by the choice of very efficient quenching agents, of high quantum stopping power. Figure 21 was obtained with a point counter filled with an argon-methyl alcohol mixture at atmospheric pressure and operated at different electrode distances.

It may be indicated here to say a word about the importance of the plateau. In the ordinary Geiger counter a plateau is desirable because working in the plateau region assures reproducibility of the counting rates even if the experimental conditions within the counter change, i.e. either if the applied voltage varies, or if the quenching gas is adsorbed or broken down by the discharge. A good plateau, therefore, can be an indicator for the life expectancy of a counter. However, in taking photographic pictures with the counter, long life of the counter is not required, and a good plateau is not needed, though it can be helpful in making the voltage (or distance) adjustment less critical. Regardless of the ultimate requirements, an investigation of the plateau is of interest since it furnishes some indications as to the mechanism of the counter action.

d. Influence of the quenching agent

An increase in the amount of quenching agents has the following effects. First, the curves are shifted to the right, i.e. to higher voltages.

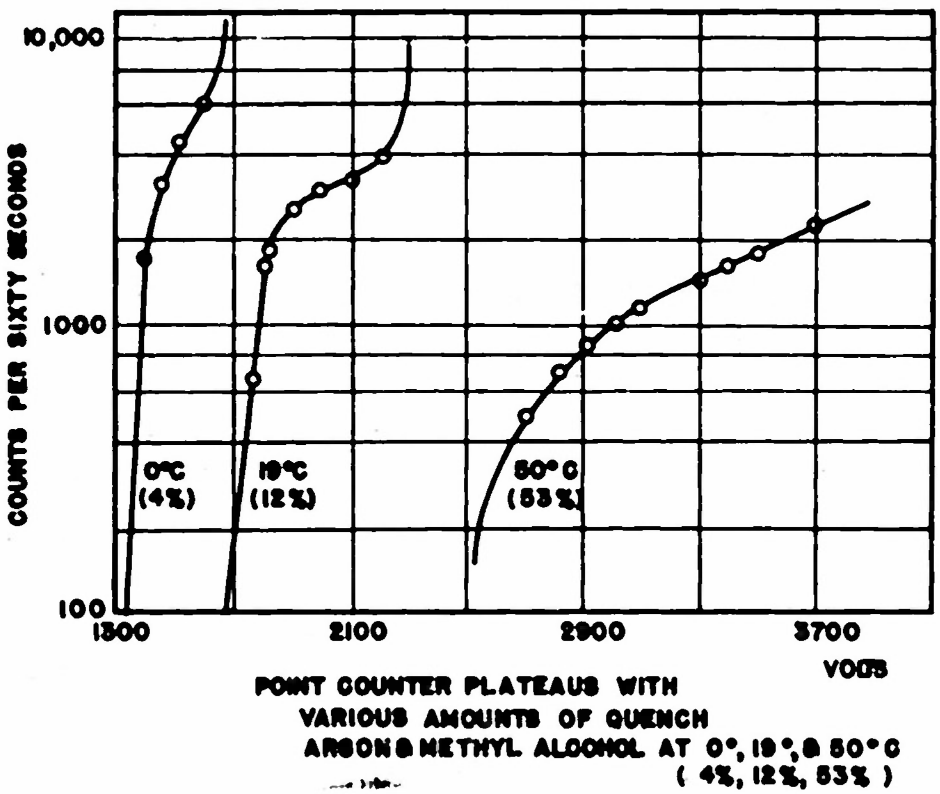


Fig. 22

This is in accordance with the general observations on Geiger counters. The presence of a polyatomic constituent in the gas raises the threshold voltage because a large portion of the electron energy is dissipated in exciting molecular vibrations of the quenching agent at each impact. An electron therefore loses energy at each inelastic collision, and is not likely to acquire the energy necessary for ionization, and higher voltages are required to enable the electron to accumulate sufficient energy over its free path. Second, the plateau becomes longer and flatter. This is to be expected from conventional Geiger counter theory. The usual explanation is that the quenching agent in some way prevents the positive ions from reinitiating a discharge when they reach the cathode. Third, the curves are shifted downward, the number of counts for any applied voltage is considerably lowered, and also the number of counts in the center of the plateau (point of inflection) is lowered, even though this point occurs at higher voltages. A possible explanation for this effect is that an increasing fraction of the initially formed electrons are captured by inelastic collision with the polyatomic constituents and are lost for the formation of avalanches. Such a reduction in quantum efficiency has been reported in ordinary Geiger counter work of cylindrical geometry.[47]

Several quenching agents were tried, such as methyl alcohol, ethyl alcohol, ether, butane, ether plus alcohol, and some others. Their effect was found to be qualitatively similar, although there seem to be pronounced quantitative differences between these agents. For instance, ether seems to be considerably more effective in extending the plateau and suppressing spurious counts than alcohol. On the other hand, some of the quenching agents other than alcohol produce very satisfactory photographic effects, but do not permit quantitative reproducible measurements. They evidently lead to the formation of discharge products and contamination of the electrodes.

e. Influence of source intensity

Figure 23 shows two curves in which the counting rate is plotted vs. the applied voltage for two different intensities of incident radiation varying by about a factor 10:1. At low voltages these two curves are close together, presumably because the influence of the large background counting raises the level of counting rate relatively higher for the low incident intensity. At higher voltages, the counting rate for both curves remains approximately at a ratio of 10:1, as would be expected.

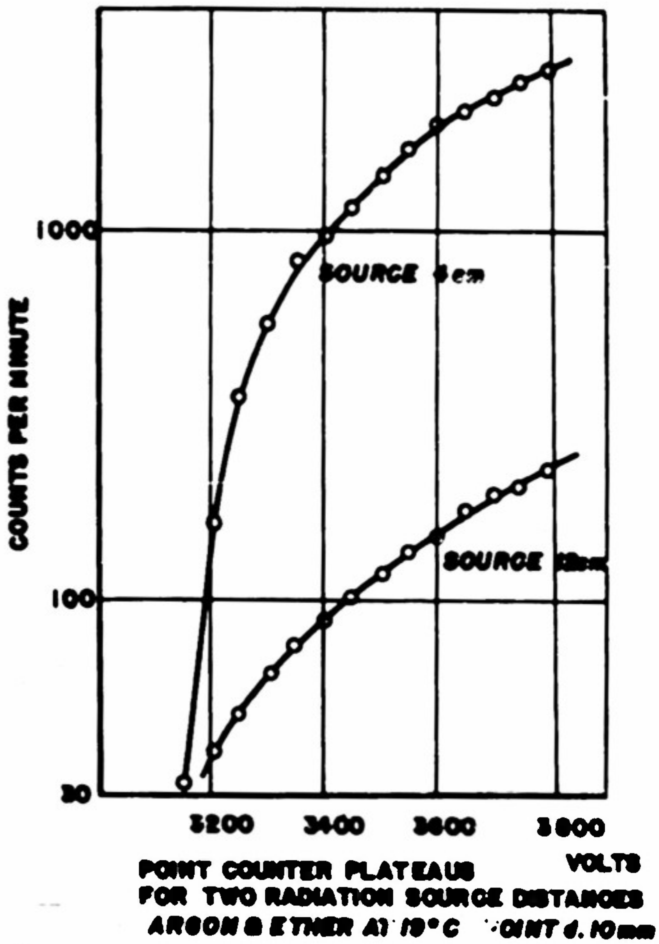
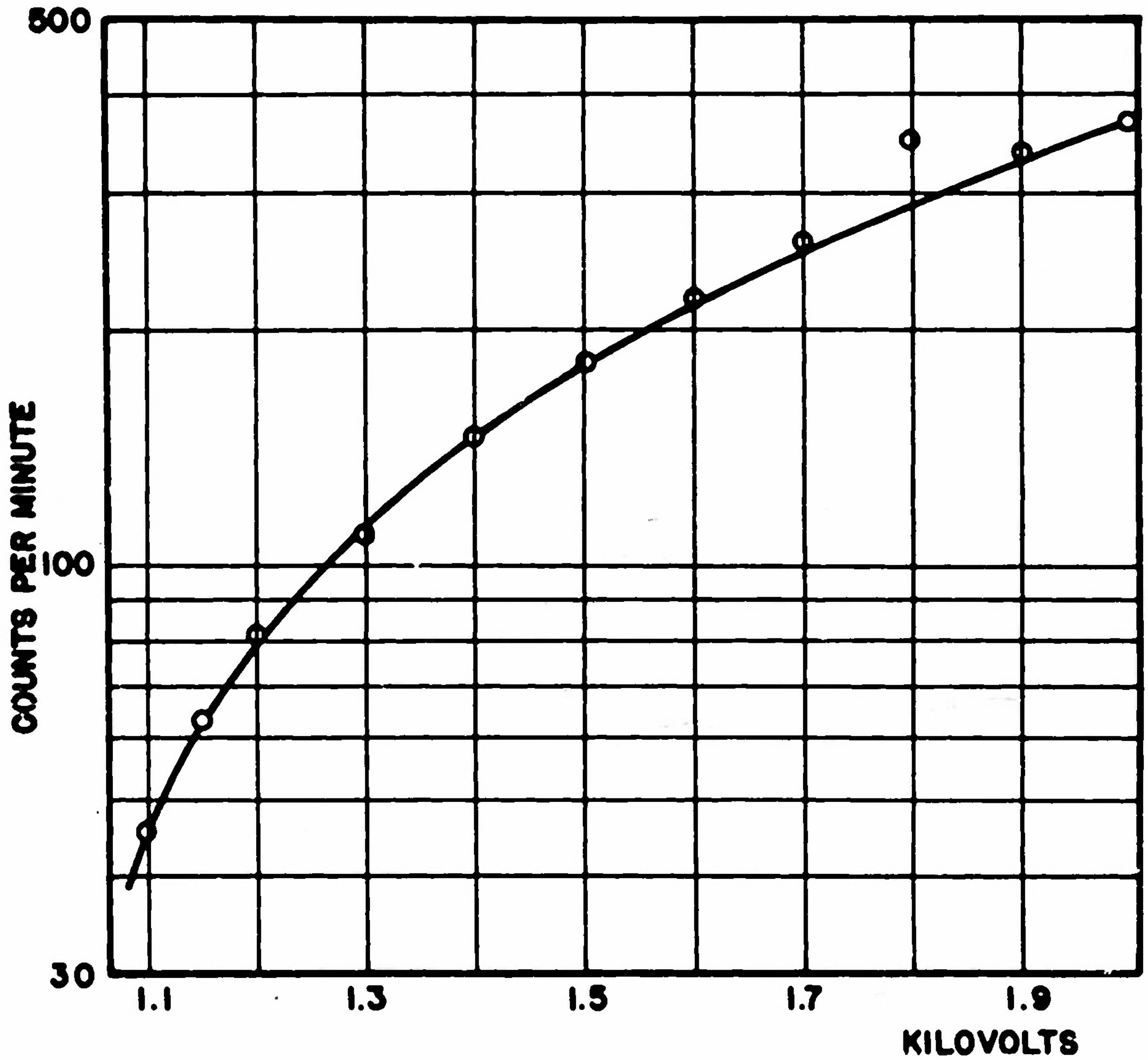


Fig. 23

f. Influence of an inserted film

When the cathode is covered with a photographic film or paper, or an insulator or semiconductor, the general behavior of the counter is more or less unchanged, indicating that within a certain range of operating conditions the cathode is not primarily of influence upon the counter action, or that the surface properties of the investigated material is similar to the one of a metal cathode (as far as electron emission is concerned). Proportional pulses appear at some voltage, and Geiger pulses follow at a higher voltage. No sparks are observed, however, indicating that the high resistance backing cf the film limits the amount of charge which can enter into any one discharge. This is also evident from the fact that the Geiger pulses observed in a counter with an insulating surface are smailer than the one in a counter without insulator. The steeply ascending branch C in the counting rate-vs-applied voltage curves (Fig. 20) is absent, and the counter characteristics tend to flatten out, an effect which is probably due to the fact that the high resistance in these commercial photographic materials slows down the recharging of the surface, so that a saturation effect results. Figure 24 shows a typical counting rate characteristic of a point-to-plane counter with positive point and an insulator as cathode.



POINT COUNTER PLATEAU WITH INSULATED CATHODE
d: .13 mm. ARGON - ETHER AT 19°C (55% ETHER)

Fig. 24

g. Photographic characteristics, general behavior

Two factors are of predominant interest in the investigations of the photographic effect of counter discharges, the local density produced in an emulsion by the discharge, and the lateral extension of the trace produced by a single discharge or by a burst of discharges upon the film. The former effect reveals the gain obtained, the latter one furnishes information as to the resolution of a picture that can be expected.

The intensity of the blackening produced by a discharge and the lateral spreading depend upon a number of physical factors. In the point-to-plane discharge it depends first upon the number of pulses passed from the point to the film, and upon the point-to-plane distance. This relationship is shown in Fig. 25, page 56, for a variety of conditions.

Both the density and the lateral extension increase with increased number of discharges. With increased distance between point and film the density falls off, and the lateral extension of the blackening increases, the trace becomes diffuse; the definition decreases moderately at point-to-plane distances between .07 and .12", and falls off sharply beyond .12". (The reverse effect in the third vertical column is probably due to a secondary effect such as higher humidity of the paper in the case of 1000 counts, .1").

The traces of Fig. 25 were produced on Kodabromide paper directly. The applied voltage varied between 2600 and 3000 volts, so as to produce satisfactory discharge pulses of the self-sustaining type. All discharge traces were produced with large pulses of the Geiger pulse type. Experiments to produce traces with the small pulses of the proportional type were unsuccessful in these experiments as in all others.

The picture of discharges shown in Fig. 25 in the left vertical column do not represent the smallest traces obtained in our experiments. Extremely small pictures have been received (depending upon the character of the discharge), but these can only be observed under the microscope. Also, depending upon the discharge character, the lateral extension of the discharge does not always increase with the number of discharges. Figure 26, page 57, shows a group of four discharge figures obtained with increased numbers of counts in the ratio

1:2:3:4, in which the effective diameter of the cluster of grains does not show an increase.

It seemed of interest to obtain by photometric analysis some data of the lateral extension of the dot, and of the local density distribution within the dot. Such measurements were carried out in the following way. A trace was produced on Commercial Ortho film by the passage of about 10 discharges in the point counter. The film was then processed in the usual manner, and projected through a microscope on a screen. A photometer head with a slit opening was moved along the screen, and the local light intensity was measured.

The resulting curve is shown in Fig. 27 together with the dimensions of

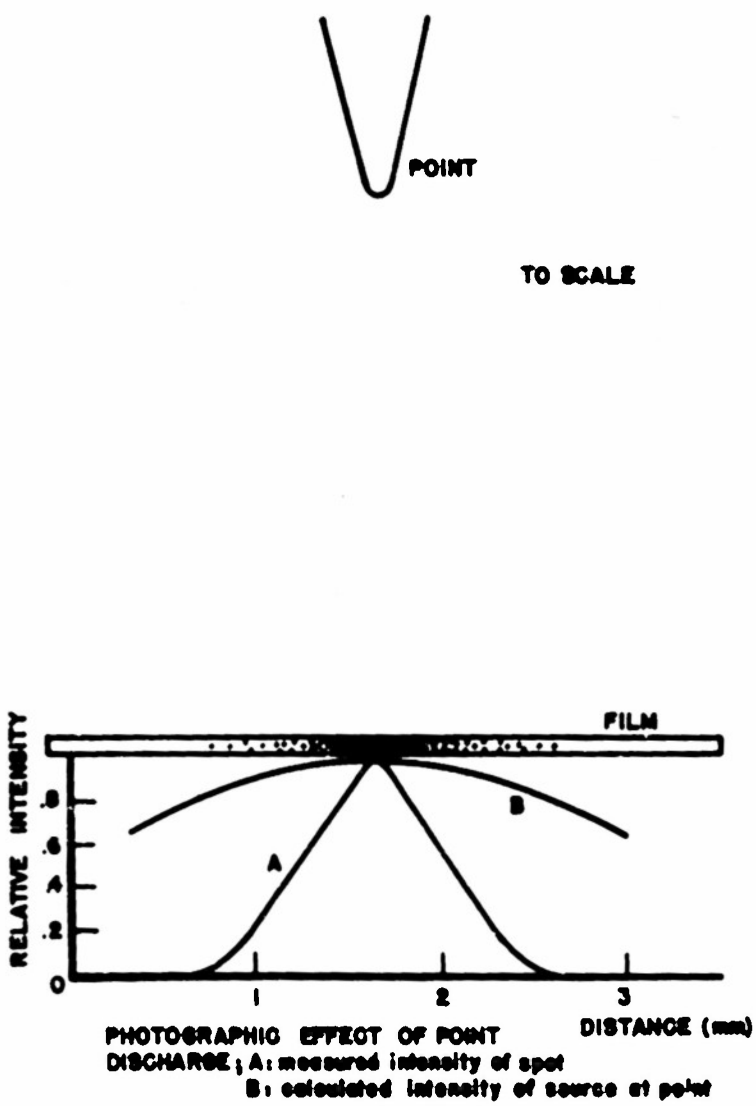


Fig. 27

the counter. It appears that the dot thus produced (density distribution, curve A) has an effective diameter of less than half the distance point-to-plane, and a very sharply marked peak density. This result excludes the possibility that the dot be produced by a corona discharge around the point (e.g. light originating at this point). The distribution of illumination on the film surface in this case would have followed an inverse square law, and the density distribution within the spot (assumed linearity between illumination and density) would have been the one indicated by the curve B.

h. Influence of film resistance

The blackening obtained by the discharge on a photographic emulsion is undoubtedly strongly influenced by the electrical resistance of the backing. This was shown in an experiment where a piece of Kodalith stripping film was removed from its celluloid backing, mounted on a metal plate and inserted into a point-to-plane counter. The needle was adjusted at a distance of .098 inches from the film and 19 large counts of the self-sustaining type were allowed to form. After processing, the film was removed from the metal plate and mounted on a microscope slide. Figure 28, page 57, shows an enlargement (35 times) of the obtained discharge figure on the film.

The experiment was then repeated with a piece of stripping film from which the celluloid backing had not been removed: under the same conditions 19 large type pulses did not produce any visible trace on the emulsion.

The fact that the resistance of the photographic paper and the emulsion influences the discharge character to such a great extent introduces a considerable experimental difficulty into our investigations. The resistance of the photographic material varies drastically with humidity over several orders of magnitude. While this has given us in the past at least a means of varying the conductivity of the film (by conditioning it in atmosphere of appropriate humidity), the effect is rather unreliable, subject to gross variations with the least amount of impurities, and varying with the age of the material, its past history, its temperature, etc. It seems that no other single factor contributes so much to the variations in the outcome of our experiments as this factor of the conductivity of the film.

Various techniques are contemplated to overcome this difficulty in our experiments. The effect of the high resistance of the emulsion carrier may be eliminated by applying an emulsion directly to metal or on semiconductors. A number of semiconducting plastic samples have been prepared for us by a commercial company (Emerson and Cuming, Canton, Mass.). The use of glass with surface conductivity or with conduction through the body of the glass by ionic conductivity with alkali ions has been considered and some information on this matter has been gathered. Also some preliminary experiments have been carried out with Du Pont "DH" dehydrated photographic emulstion and with strip film, applied to metal plates. The use of emulsion pellicules for the same purpose is planned. The resistance of the emulsion may be controlled by impregnation with different salt solutions, such as KCl and others. Experiments carried out so far have been encouraging.

i. Penetration depth in the emulsion

In an attempt to obtain some further information about the origin of the photographic effect, we set out to observe the penetration depth within the photographic emulsion. It was expected that observations of this kind would give us information as to the kind of radiation or particles which produced the blackening effect upon the photographic film. If light had caused this blackening, there would be exposed grains throughout the cross section of the emulsion. An exposure of the grains produced by electrons, ions, and extreme ultraviolet would be confined to the surface of the emulsion.

Ten discharges in a point-to-plane counter in air at atmospheric pressure were produced on an Ortho-X film. The film was processed in the usual manner, dried and imbedded in a paraffin block. The block was sliced with a microtome and sections of a thickness of about 10 μ were made. These sections were prepared for microscopic observations.

Figures 29a and b, page 57, show cross sections of such a film at a magnification of 100. Figures c and d show similar slices at a magnification of 450. The exposure of the grain is limited to the surface of the emulsion, but penetrates up to about two-thirds of the thickness. Such a phenomenon excludes the participation of visible light as a cause for the blackening. On the other hand, it seems unlikely that electrons of low velocity or ions could have produced the effect since their action would have been limited to the very surface. It seems likely that the blackening is produced by ultraviolet light in the range of about 300 $m\mu$.

Figures 30, a and b, page 57, show the cross section through a film exposed to light. The blackening is fairly homogeneous throughout the cross section of the emulsion. It appears that the structure of those grains exposed by the discharge is much coarser than the one produced by light.

These measurements are in agreement with others in which the depth of penetration was measured by focusing a microscope towards successive layers in the emulsion. When exposed with light, developed grains would be observed throughout a depth of 15 microns, while in emulsions exposed to discharges, developed grains were found primarily within a depth of 3 microns.

2. Parallel plane counters

The experiments on point counters had shown that a marked change in the behavior of the counter occurred when the needle was brought close to the plane electrode. At distances less than ten times the radius of curvature of the needle, the discharge form changed in a very peculiar way: The plateau became less pronounced and showed a very steep slope, and the counter discharge showed a transition with increasing voltage from proportional pulses to triggerable spark pulses when used without film. At these close electrode distances the point counter approaches in effect the parallel plane counter. It is to be expected, therefore, that also the parallel plane counter will exhibit similar characteristics as the point counter at very close electrode distances, however, with two important variations. First, the fieldstrength is uniform throughout the counter volume, at least prior to the formation of a pulse, and second, its electrical capacitance is far greater. These effects can profoundly alter the pulse character.

Reproducible measurements in parallel plane counters are even more difficult than in point counters, presumably because the very powerful spark discharges occurring in such counters are apt to alter the surface of the electrodes directly, and to disintegrate the quenching agent so that the disintegration products apparently also react with the electrode surfaces and modify their electron emission characteristics. The experimental values communicated in the following are to be understood, therefore, as qualitative illustrations of the general behavior of the parallel plane counter.

a. Discharge characteristics, general behavior

A cross section of the parallel plane counter used for these experiments in shown in Fig. 31. The upper plate consists of a piece of conductive

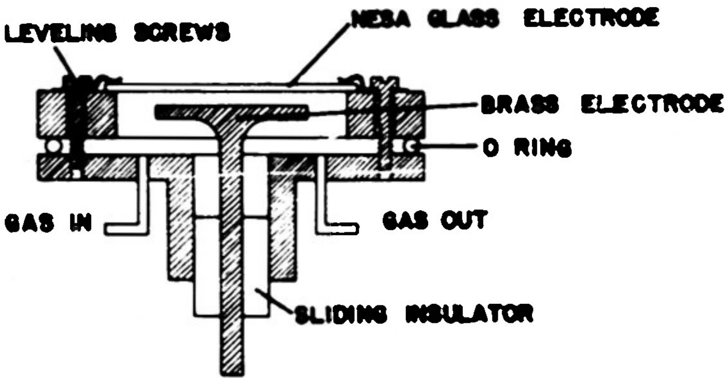
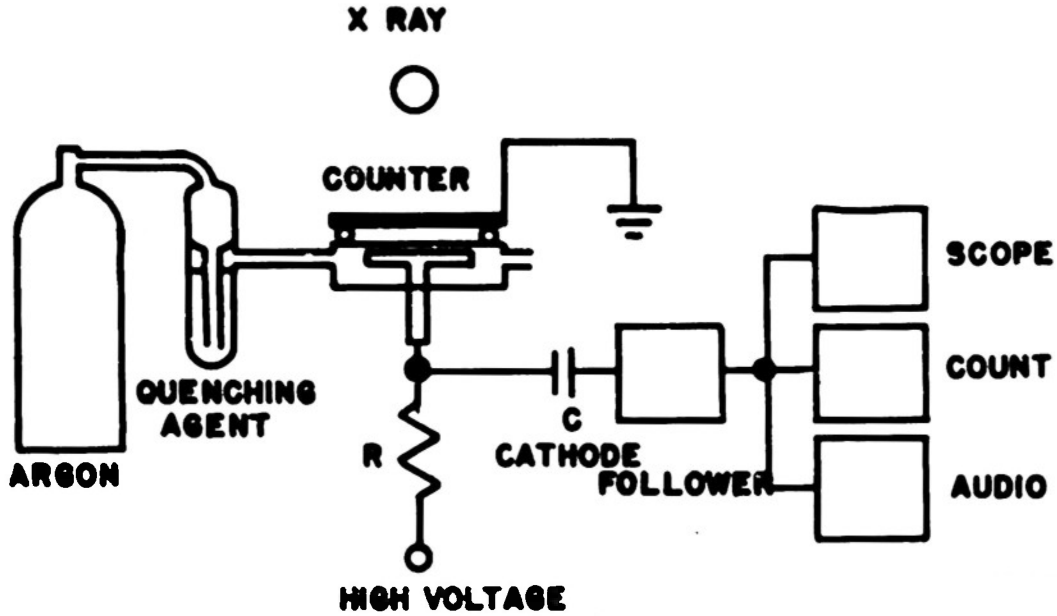


Fig. 31.

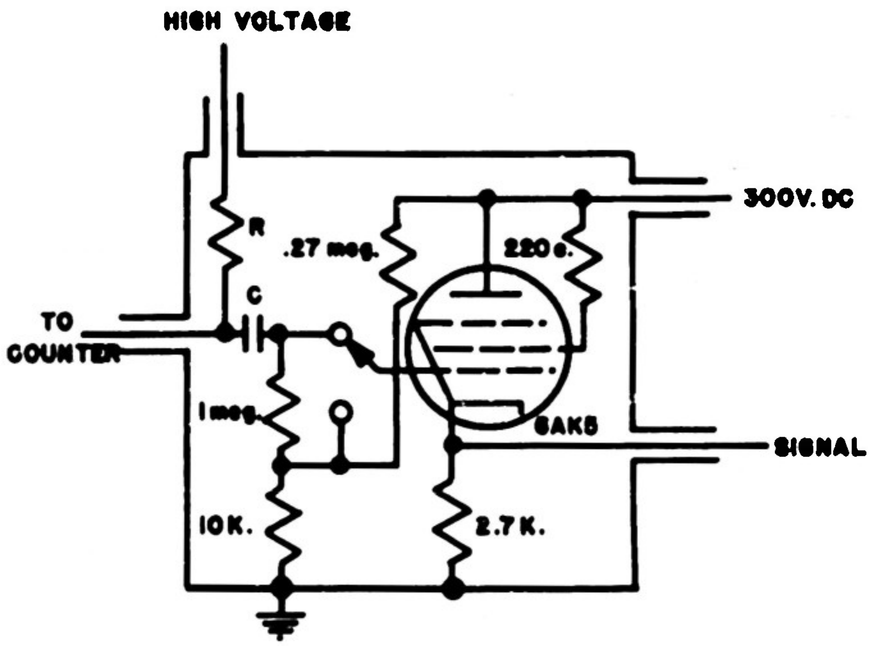
glass (NESA glass from the Libby Owens Ford Company, i.e. a glass plate with a transparent conductive surface; apparently of stannous chloride), as suggested by Johnson.[24] The experimental setup is similar to the one used in the previous experiments, and is illustrated in Fig. 32. The great variety of pulses, ranging from the order of 1000 volts down into the millivolt region, made it necessary to attenuate these pulses in the cathode follower input stage, the circuit of which is shown in Fig. 33.

The general behavior of this counter (without resistive film inserted in it) is the following: As the voltage across the counter is raised, no current flows through the gas gap until at voltages in the vicinity of 1000 volts small (10 mV) proportional pulses begin to appear. These pulses increase rapidly in size with increasing counter voltage until at some well defined voltage Geiger



PLANE PARALLEL COUNTER SET UP

Fig. 32



CATHODE FOLLOWER & ATTENUATOR

Fig. 33

pulses appear. In the parallel plane counter with noble gas filling and with a small admixture of a quenching agent these Geiger pulses appear as small intense sparks corresponding to a pulse of voltage almost equal to the applied voltage. (~1000 volts). The sparks are triggerable and stable over a certain sharply rising plateau region at the end of which the counter breaks into continuous arc discharges which tend to deteriorate the electrodes.

The results of these measurements are shown in Fig. 34, and were obtained at a distance of .008 inches between the electrodes, the counter being

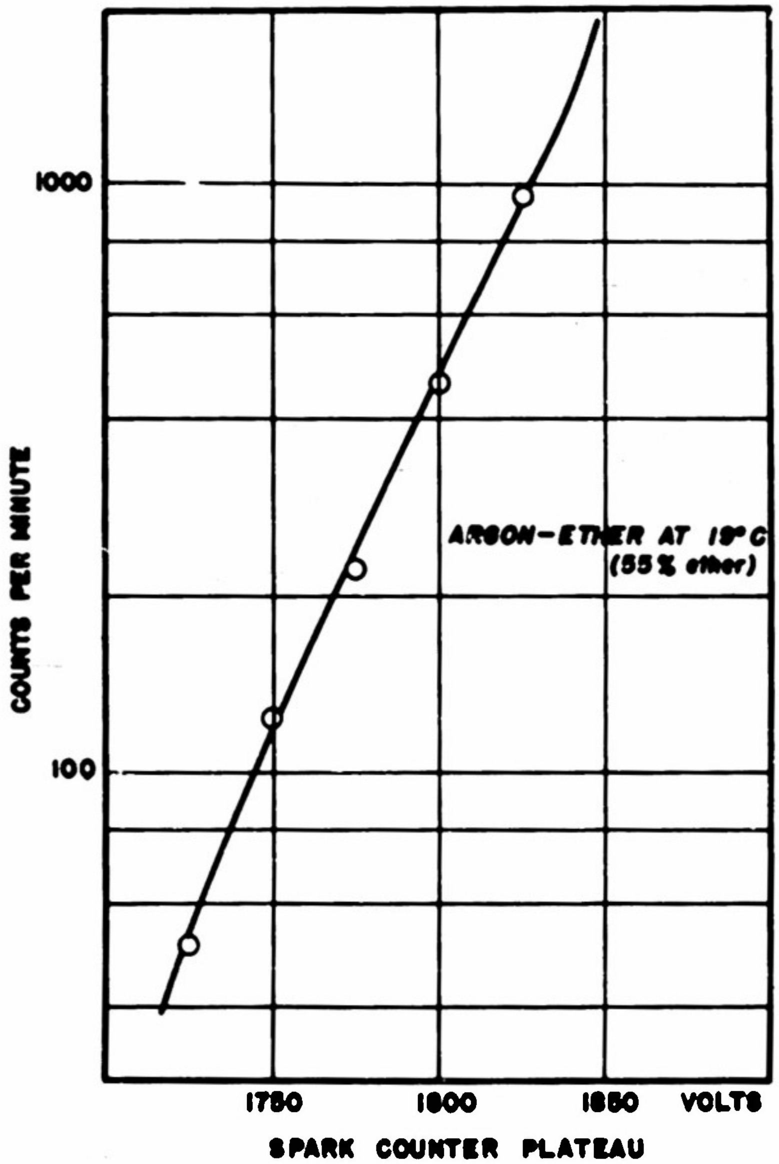


Fig. 34

filled with a mixture of argon with ether. The plateau curve was taken by increasing the voltage in steps of about twenty-five volts. A radium source at a fixed distance from the counter provided sufficient incident radiation. The same procedure was repeated twice more. The resultant curves show a pronounced drift towards higher voltages. The experiment was repeated using an aluminum electrode instead of the NESA glass, and also using alcohol instead of ether as quenching agent. In both cases the plateau was observed to drift. The general steeply rising plateau characteristic may be noted, however.

The difference in the behavior of the parallel plane counter as compared with the point counter seems to be due to two factors: the difference in field configuration and the difference in electrical capacitance.

An ordinary point counter operated at large electrode distances and filled with an argon alcohol mixture is self-quenching. It is commonly assumed that the discharge in such a counter is terminated by the following mechanism: A positive ion sheath surrounds the anode and increases in radius, thereby increasing the effective radius of curvature of the anode. The fieldstrength surrounding the ion sheath decreases, therefore, and the point is soon reached where the fieldstrength falls below a certain critical value where a discharge cannot further be maintained. The discharge then ceases, the positive ions drift towards the cathode, and the quenching agent prevents the ions from initiating new avalanches. Now, if the positive point is brought in close proximity to the cathode, the space between the electrodes is divided into two parts with the positive ion sheath between them. The fieldstrength between the positive space charge and the anode is reduced, but the fieldstrength between the positive space charge and the cathode is now increased. Further ionization and avalanche formation is then possible, and an external mechanism is required to quench the discharge. This is the case in the point counter at close electrode distances, and even more so in the parallel plane counter. The termination of the discharge is brought about through the voltage drop in the external resistor and the corresponding decrease of the voltage applied to the counter below the minimum sustaining voltage. If the capacitance between the electrodes is small (point counter at large distance), only a small charge is available for the discharge process. The voltage available to maintain the fieldstrength for the discharge mechanism will drop rapidly after the initiation of the discharge before the pulse has grown extensively, and the pulse height in such a counter will be small. A capacitor connected in parallel to the discharge path (see Fig. 13) will have the effect that the voltage between the electrodes will remain high enough for a sufficiently long time so that the pulse can grow and eventually develop into a spark. This, then, is the mechanism in the Greinacher and Rosenblum spark counters. Termination of the discharge occurs when the capacitor is discharged to a voltage level at which the discharge cannot be maintained any longer. This level is lower for the spark discharge than for the Geiger pulse, and the pulse size in spark counters is therefore very high.

The two effects of the close distance between the electrodes and of large capacitance are obviously superimposed in the parallel plane counter. The steeply rising plateau characteristics and the large pulse size can thus be understood. However, the mechanism in a parallel plane counter is most probably more complicated and an influence of the quenching agent (which is not mentioned above) is clearly noticeable.

b. Influence of the electrode spacing and the quenching agent

Plotting of the threshold voltages at which Geiger (i.e. spark) counting begins in a parallel plane counter versus the separation distance of the electrodes reveals a strict linearity. This fact indicates that the only requirement for Geiger pulse formation in such counters is the presence of a certain critical fieldstrength, as is expected in the homogeneous field of such a counter. The minimum fieldstrength varies, of course, for different gases and different admixtures of quenching agents, as is shown in Fig. 35 for argon-ether (44,000 Volts/cm) and for argon alcohol (18,300 V/cm). The threshold for pure argon is practically identical to the curve for alcohol quench, indicating the greater efficiency of ether over alcohol as a quenching agent.

However, it must be understood that there is still a considerable difference in the behavior of the counter when all quenching is removed. Even though the threshold may not be effected, the counting rate in pure argon rises with extreme steepness as the overvoltage is increased, tending to be almost completely

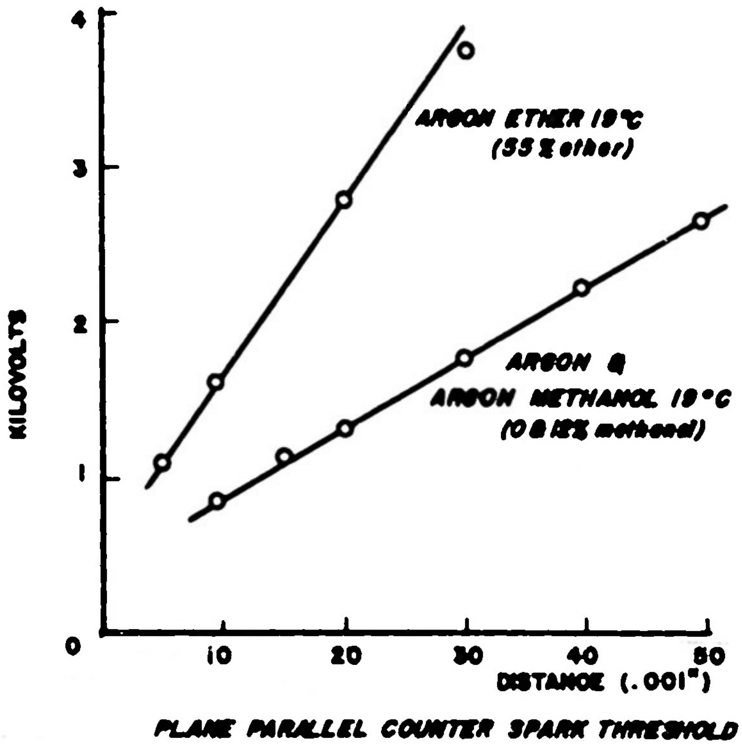


Fig. 35

nontriggerable and lapsing almost at once into arcing.

As mentioned before, the pulse size in such counters (without the presence of a photographic material) is enormous, and Fig. 36 shows a graph of pulse size versus electrode distance. The voltage for each distance is adjusted to the Geiger threshold level in this graph.

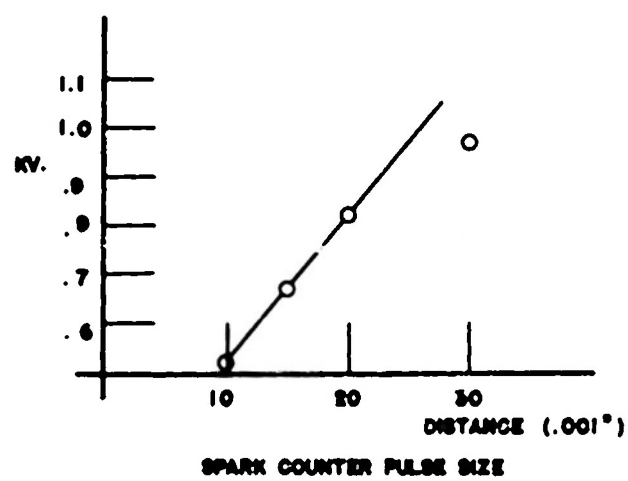


Fig. 36

c. Discharge localization

Of particular interest in the application of the parallel plane counter for photographic purposes is the question of the localization of the discharge. Obviously, a counter would be useless for the intended purpose if discharges would occur in places other than those where the incident radiation strikes the counter or, if a spark would spread within the counter, or would trigger other sparks in some remote part of the counter.

To prove that none of such effects occur, the following experiment was carried out. A photographic film was mounted outside of the counter, but in direct contact with the NESA glass window. A lead figure was placed upon the back of the film and X-rays were permitted to enter the counter through the film. The arrangement is shown in Figs. 37, a and b. At the places where the incident X-rays triggered the discharge, sparks were formed which produced a picture on the photographic emulsion, while at the place where the lead cross prevented the X-rays from entering the counter, no such effects occurred. The

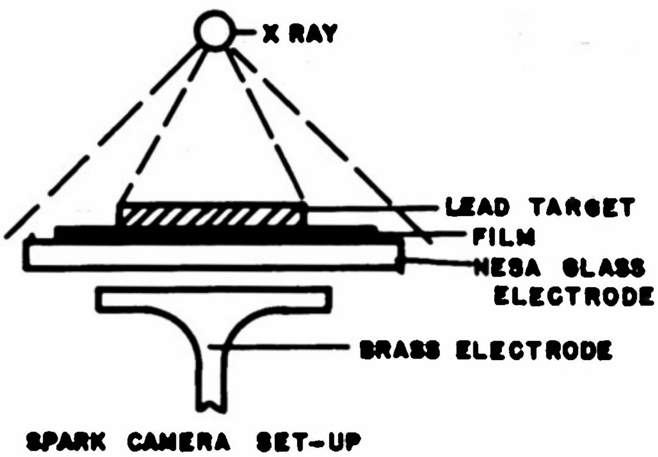


Fig. 37

obtained picture which is reproduced in Fig. 38, page 57, shows the result. The ill-defined outline of this figure is, of course, the result of the separation (2 mm) between the photographic film and the discharge space. The total exposure time for this picture was only two seconds during which time 220 pulses were formed.

Although X-rays and gamma radiation from Ra and Co^{60} were used in these experiments, it was found that discharges could be initiated with ordinary visible light of sufficient intensity. This was particularly the case when the brass electrode was made negative so that electrons ejected from it by photoelectric effect would be accelerated by the field and initiate a discharge.

d. Influence of an inserted resistive film

The insertion of a resistive film into a parallel plane counter does not appear to effect the threshold fieldstrength for the occurrence of Geiger pulses. As Fig. 39 shows, this fieldstrength remains in the order of 40,000 V/cm in a counter filled with an argon ether mixture. However, no sparks occur when

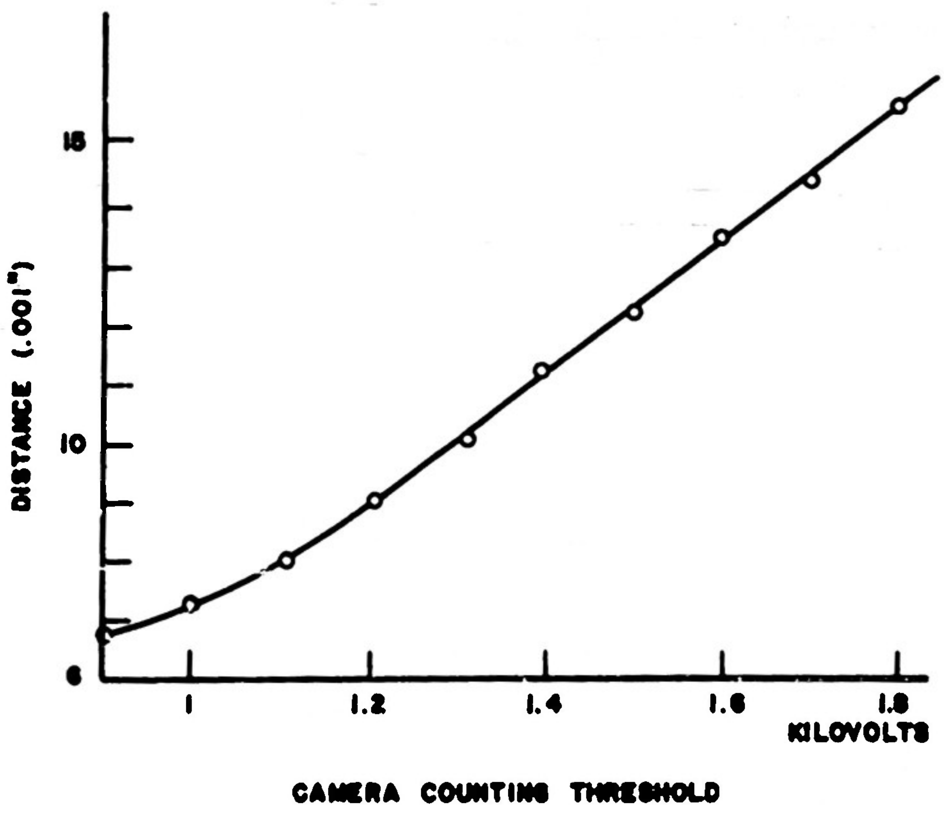


Fig. 39

a resistive film is inserted in the counter, and the pulse size is greatly diminished, varying between 1 and 10 volts as shown in Fig. 40.

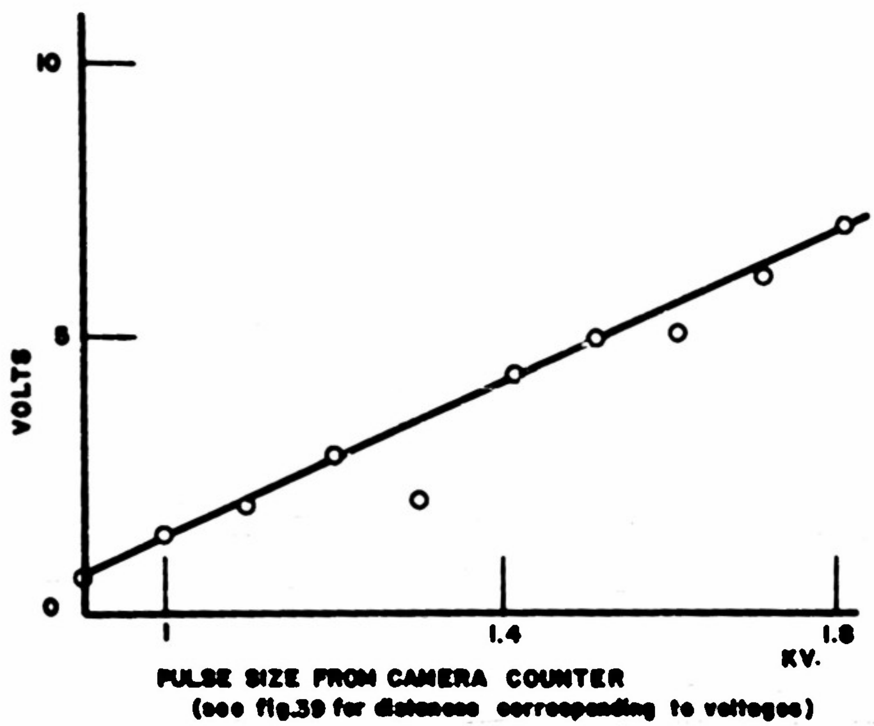
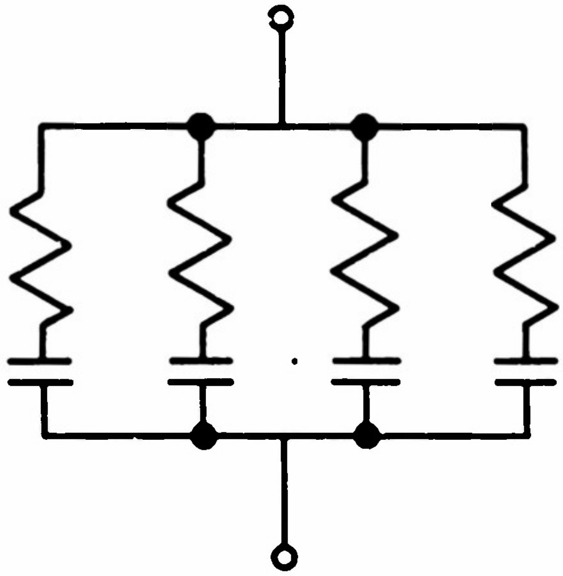


Fig. 40

The different behavior of a counter with a resistive film seems to be of a purely electrical nature. It has been pointed out, above, that the parallel plane counter is not self-quenching, but requires the presence of a resistance in the connection between the counter and the voltage source to terminate the discharge. For a distance setting around .01 inches the counter discharge will break into an arc if the external resistance is less than one megohm. The resistive film in the counter furnishes the required quenching resistance, but also changes the effective capacitance of the counter. This may be explained with the help of the equivalent circuit, Fig. 41. As long as no discharge occurs, the surface of the film will assume the potential of the electrode to which it is attached. Any discharge will then take place between the surface of the resistive film and the opposite electrode. The film backing, thereby, acts as a quenching resistance. However, the surface area involved in a discharge is small, and, therefore, forms only a small capacitor which is effectively isolated from a neighboring area or



RC NETWORK

Fig. 41

from the voltage supply. The total amount of charge available to produce a discharge current high enough to characterize a spark is not available, therefore, and the discharge character, as far as the pulse size or current density is concerned, is similar to the one of a point counter.

Obviously, it should be possible to influence the discharge character in such a counter profoundly by variation of the resistance of the inserted film. Such experiments are planned as one of the next steps in this project, in connection with the investigation of the photographic effect of discharges in a parallel plane counter (see next paragraph).

e. Photographic effects in parallel plane counters

Two cameras were developed for the investigations of the photographic effect on parallel plane counters. Figure 42, page 58, shows a double drum camera for series of exposures on film or paper that can be moved through the camera without opening it and renewing the gas filling. Figures 43, a and b, page 58, show a camera for single exposures. This camera is intended for precision work and permits fine adjustment of the electrode spacing and the electrode parallelism.

Only orientating experiments have been carried out so far with both instruments. One of the results of these experiments is shown in Fig. 44, page 59. These pictures were taken in the drum camera with a spherically shaped anode in an atmosphere of nitrogen and ether. The setup for the 10 different pictures is given in Table II. One can easily recognize that the blackening increases with the pulse size.

Independent of these experiments, pictures were taken with different experimental setups, and some of the results are shown in Fig. 45, page 59.

TABLE II

No.	Volts	Distance (Thousandths of inch)	Pulse Size
1	900	6.50	1 V.
2	1,000	7.25	1.5 V.
3	1,100	8.0	1.8 V.
4	1,250	9.0	3 V.
5	1,300	10.0	2.6 V.
6	1,400	11.25	4.4 V.
7	1,500	12.25	5 V.
8	1,600	13.50	5 V.
9	1,700	14.50	6 V.
10	1,800	15.75	7 V.

Photographic material. Kodabromide, F3

Gas: N_2; Quenching agent: Ether @ 19°C (55%)

X-ray setting: 87 volts

Film humidity: 56%

Number of pulses for each exposure: 2,048.

E. CONCLUSIONS

It must be assumed that in the initial steps of the discharge formation, the process in the parallel plane counter with resistive film is similar to the one in the conventional cylindrical Geiger counter. However, a different mode of operation apparently sets in when the head of the avalanche reaches the anode. It has been shown by numerous experiments that in the cylindrical counter the discharge spreads in a narrow sheath along the anode. In the parallel plane spark counter, no such spreading has ever been observed; the discharge remains rather strictly localized, as indicated by the observation of the spark and by the appearance of the photographs taken with the counter. These photographs show excellent localization, regardless of whether the photographic emulsion was attached to the cathode or the anode. Although Raether[48] has shown that the development of the avalanche into the spark canal remains strictly localized, and that even a radial electric field arises within each canal which concentrates the discharge, it is questionable whether his reasoning is applicable to the situation in parallel plane counters with resistive film. The investigation of this problem as well as experimental studies on the photographic effects in the parallel plane counters are the problems to be dealt with as the next step in the present project.

It should be emphasized that the authors of this report are aware of the hypothetical character of the description of the counter mechanisms at the present time, a d of the rather qualitative character of some of the presently described investigations. Yet, it seemed advisable to obtain first an over-all picture of the processes going on in a counter and leading to the formation of a picture, rather than to spend time and effort on the exact measurement of any one detail which may turn out later to be of little consequence in the operation of the system. On the other hand, the photographic recording in the counter offers a unique possibility to obtain valuable information not only for the practical application of the method of electronic photography, but also of possible value for a general counter theory.

A great number of topics of considerable interest in connection with the electronic photographic method have not been investigated as yet. The method at the present time still uses the external photoelectric effect on an unprepared electrode surface for the liberation of electrons and the initiation of the discharge. Replacing this surface by a combination of a fluorescent and a photoemissive layer as used by Coltman[3] should increase the gain of the method by a factor of a thousand. The use of higher pressure in the counter, of a crystalline medium instead of a gaseous discharge medium, the use of special emulsions or of sensitized emulsions, of multiple quenching agents, and many other ideas have not been tried as yet, but some of these experiments are in preparation.

F. APPENDIX, DIRECT ELECTRICAL EFFECTS ON PHOTOGRAPHIC EMULSIONS

There is a possibility that the effect of the discharges upon the photographic emulsion is, in part, of an electrical nature. That means that neither ions, nor electrons or photons from the discharge produce an image on the photographic plate, but that the discharge produces an electric field of considerable magnitude within the emulsion by bridging the potential difference across the gas layer. Under the influence of such an electric field applied to the emulsion during exposure, effects of an unknown nature may occur which may give rise to an increased sensitivity.

From a theoretical point of view, such an effect does not seem very probable if one only considers the ionizing action of an electron liberated within the emulsion by exposure to radiation. An ionizing effect is only to be expected if the electron on its path between two collisions can accumulate enough energy to ionize. Assuming an energy level of the electron in the order of 1 eVolt, corresponding to a radiation of about 12,000 Å.U., being needed for ionization (usually 5 to 10 eVolts are required to free one secondary electron in silver halides), and assuming furthermore a free mean path of an electron within the AgBr crystal in the order of 10^{-6} cm, an applied fieldstrength of about 10^{6} volts/cm would be required which is impractical. However, the silver bromide crystal shows photoconductivity, and an exposure to light during the application of an electric field may lead to increased conductivity of those grains in which photons are absorbed, so that further grains in successive layers (along the same electric force line) may be exposed to higher fieldstrength. Whether or not this increased fieldstrength would lead to an electric "exposure" of these grains cannot readily be estimated.

An orientating experiment whereby a photographic film was brought into an electric field (electrodes in contact with the film) during exposure with X-rays showed a negative result (Figs. 46, a to d, page 60). Within the investigated range of incident X-ray intensities and applied fieldstrengths there is no influence of the applied voltage upon the blackening caused by X-rays. Only in figure 46c can one observe a corona effect along the edges of the electrode, and on some points in the center where the electrode may not have made good contact with the film. The negative result of this experiment contradicted some earlier observations where blackening of a photographic film was observed when a needle connected to a high voltage source was brought in direct contact with the emulsion while the other pole of the high voltage supply was connected to the back of the film. The point underneath the electrode was black and was sometimes surrounded by a halo of less density than the general fog level of the film. Figures 47, a, b and c, page 60, show several examples of this effect.

The blackening effects of the discharge and the effects of an electric current passing through the photographic film are superimposed in our photographic method, and it seemed of interest to separate both effects and to investigate separately the effects of an electric current upon the emulsion. In the beginning of these experiments it appeared very difficult to reproduce these current effects, and only after we had stabilized the humidity, the temperature, and the pressure with which the needle touches the emulsion were we able to obtain acceptable results.

The experiments were carried out in the following way: A photographic film (Commercial Ortho) was inserted in a light tight box and maintained at constant temperature and humidity for several hours. Two needles connected to a high voltage supply were then lowered carefully to avoid mechanical pressure or scratches until they made contact with the emulsion. Voltage was then applied and current and voltage were measured.

The general appearance of the electrical exposure figures is the following: Underneath the positive needle appears a dot which grows with increased exposure time and increased applied voltage (Figs. 48, a and b, page 60). Underneath the negative needle occurs a feathery discharge figure of an appearance similar to a positive Lichtenberg figure, (Fig. 48c, page 60). This appearance of both figures changes when the film has been pre-exposed to visible light and when the emulsion is maintained in an atmosphere of high humidity (order of 98 percent) before and during the application of a voltage. In this case the figures are usually surrounded by intense light halos in the order of 1 mm diameter. The figure underneath the positive electrode shows a complicated structure. At short exposure times, below one or two seconds; a small dot appears where the needle touches the emulsion (Fig. 49a, page 61); this dot is surrounded by a light halo. As the exposure time increases the center becomes light and is surrounded by a number of alternating dark and light rings, of which two are in the immediate vicinity of the point of contact, and a third light and dark halo is in considerable distance from the center. The size of this figure grows with increased exposure time (Figs. 49, b and c, page 61).

At the places where the light halo occurs, not only is the original pre-exposure erased, but the sensitivity of the emulsion is irreversibly destroyed.

We are inclined to ascribe these phenomena to electrolytic processes occurring within the emulsion during the passage of electric current. The effects occur only at very high degrees of humidity. The resistance between the two needles under such conditions is in the order of 10^8 ohms, and a current of several microamperes can be maintained. The figure at the negative electrode resembles the phenomenon observable in the electrolysis (silver tree, lead tree). It is likely that hydrogen ions migrating towards the cathode reduce the silver bromide to metallic silver. The migration of these ions follows a path of low resistance which depends upon the irregular distribution of water in the emulsion. Around the negative figure there is, of course, a region of increased oxygen ion density where oxidation and, therefore, bleaching and destruction of the silver bromide crystals may occur.

The situation on the positive point is different and the figure resembles the one of Liesegang's rings.[49]

The problem is not cleared up by this short investigation. However, since this investigation is only incidental to our main problem, we have not continued any work in this direction.

The fortunate result of these experiments is the evidence that the electric current effect upon photographic emulsions occurs in a range of physical conditions (current, time) which is different from the one in our present gas discharge experiments, and that this electric effect is therefore not of any disadvantageous consequence. Figure 50, page 61, shows the photographic effect of a single discharge produced under the same conditions of humidity and temperature as in the preceding figures. No halo or reduction in sensitivity can be observed in this picture or in others taken with multiple discharges.

ACKNOWLEDGMENT

We are particularly fortunate in that our work has found the interest, the encouragement, and the active help of such outstanding specialists in the field of electrical gas discharges as Dr. A. von Engel in Oxford, and Dr. S. C. Brown of M.I.T. At the invitation of Dr. von Engel, K. S. Lion spent a most profitable time in the Clarendon Laboratory in Oxford, and had an opportunity to discuss this project and others related to it with Dr. von Engel and his excellent staff. Dr. S. C. Brown has spent many hours discussing and advising us both in our experimental work and in the interpretation of our results. We are also indebted to Professor Allis for his interest and for numerous suggestions.

Fig. 4
from Mayneord and Belcher

Fig. 10 Lichtenberg figures

Fig.12 Discharge figure
of the Gemant type (mica)

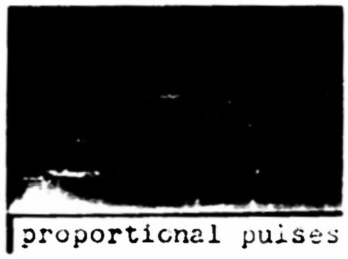


Fig. 17a

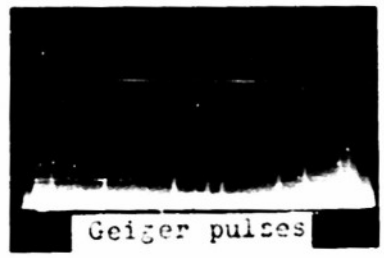


Fig. 17b

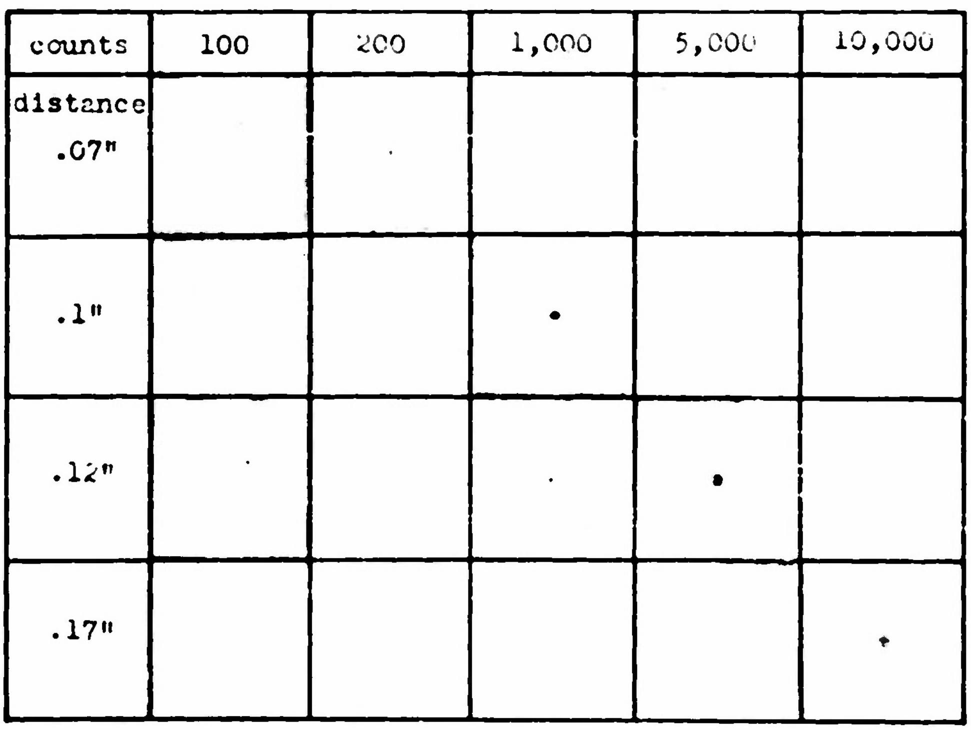


Fig. 25 Point counter discharges

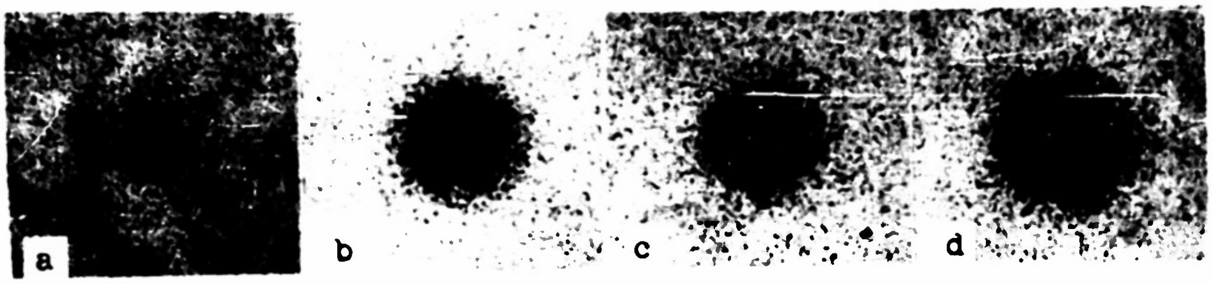


Fig. 26 Single and multiple discharge figures

Fig. 28
Kodalith film on metal base

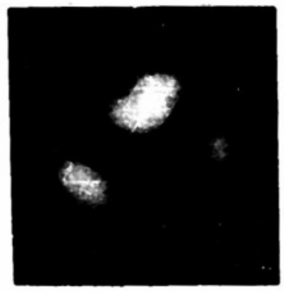


Fig. 38
Spark counter exposure

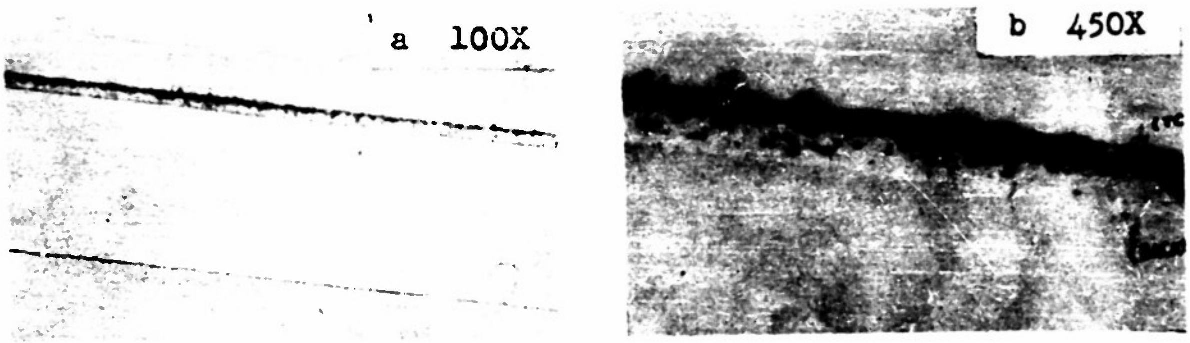


Fig. 29 Cross section through film exposed to discharges

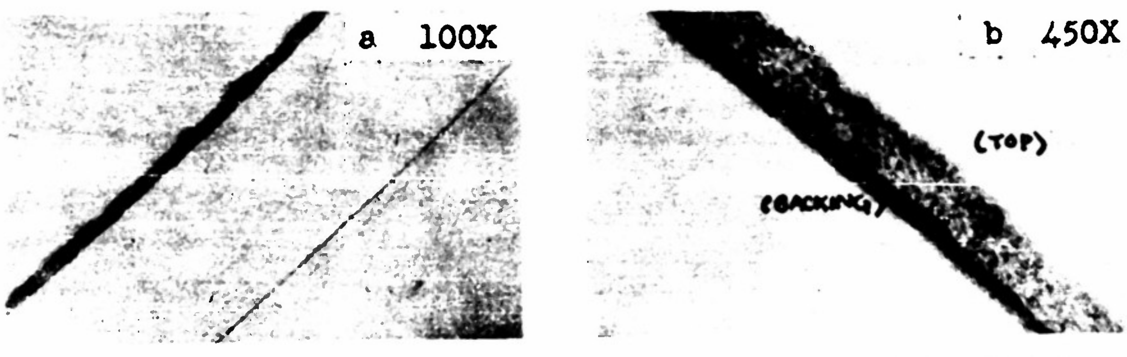


Fig. 30 Cross section through film exposed to light

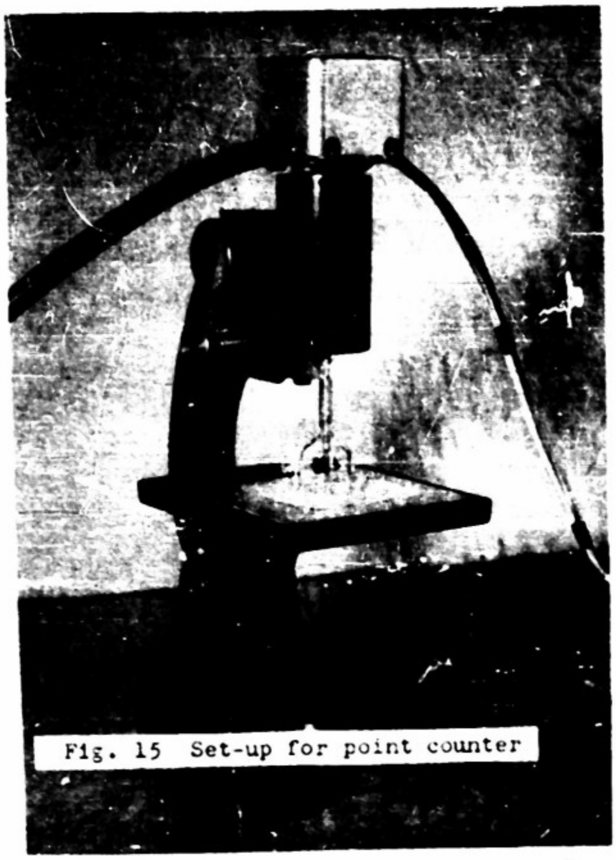


Fig. 15 Set-up for point counter

Fig. 42 Drum camera

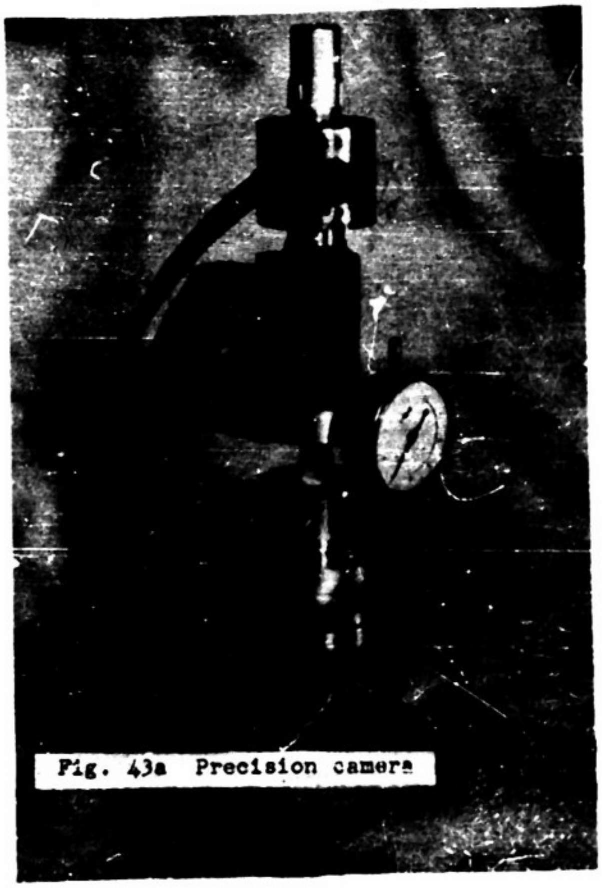


Fig. 43a Precision camera

Fig. 43b Precision camera film holder

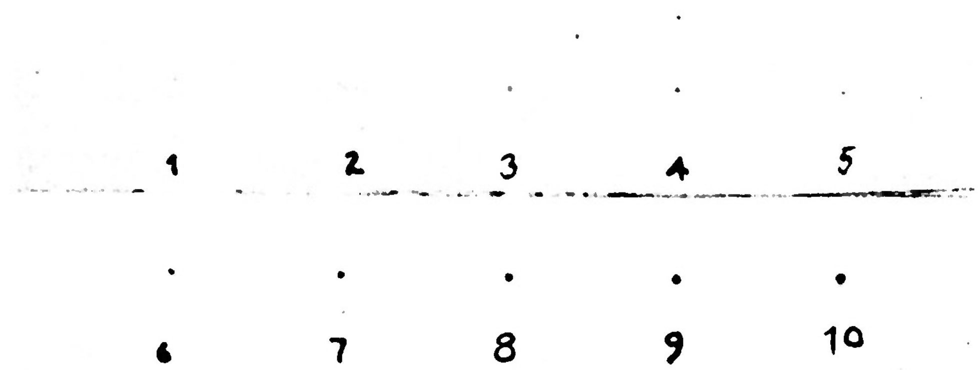


Fig. 44 Photographic effect in plane parallel counter

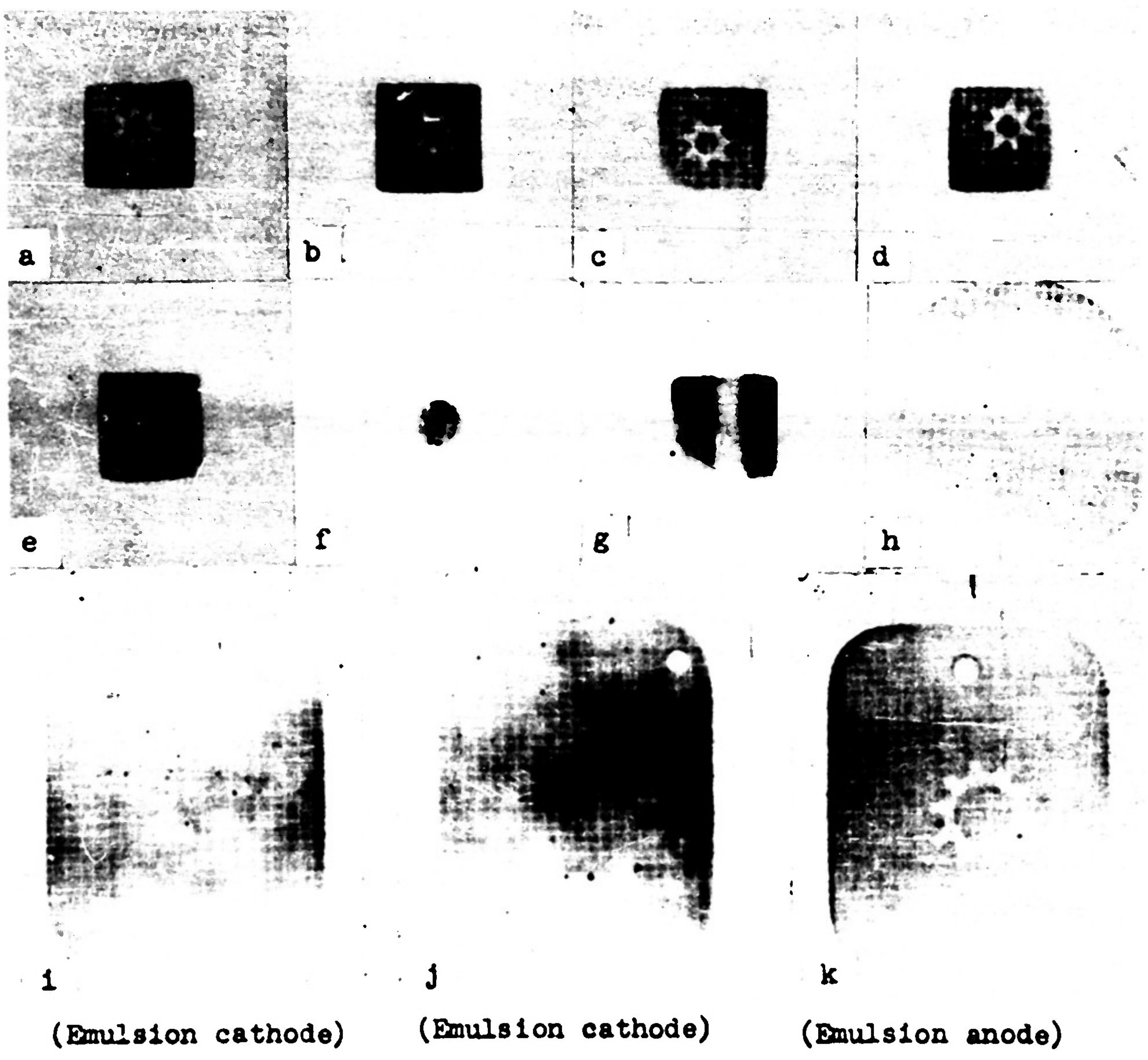


Fig. 45 Radiographs taken with different settings

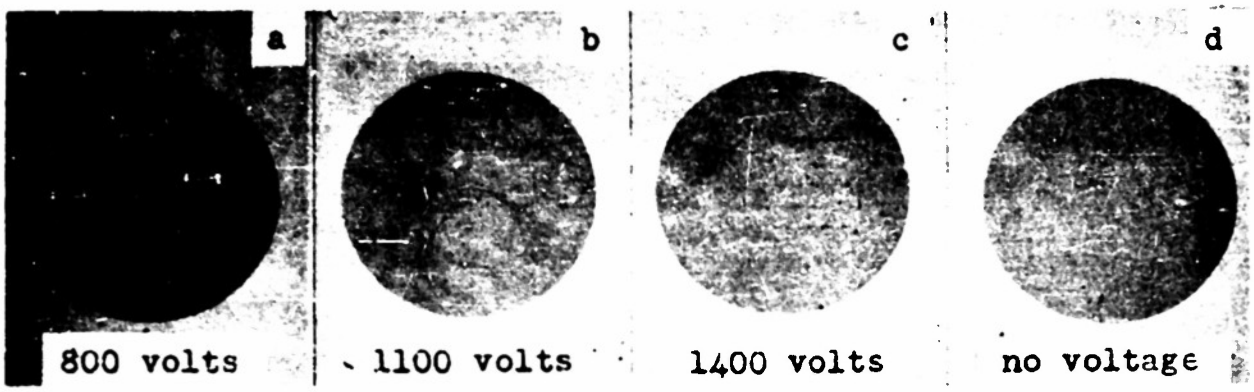


Fig. 46 Electrodes in direct contact with emulsion

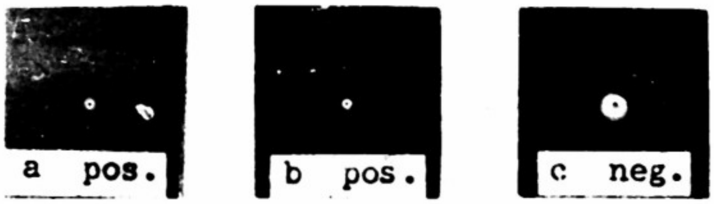


Fig. 47 Needle electrodes in contact with emulsion

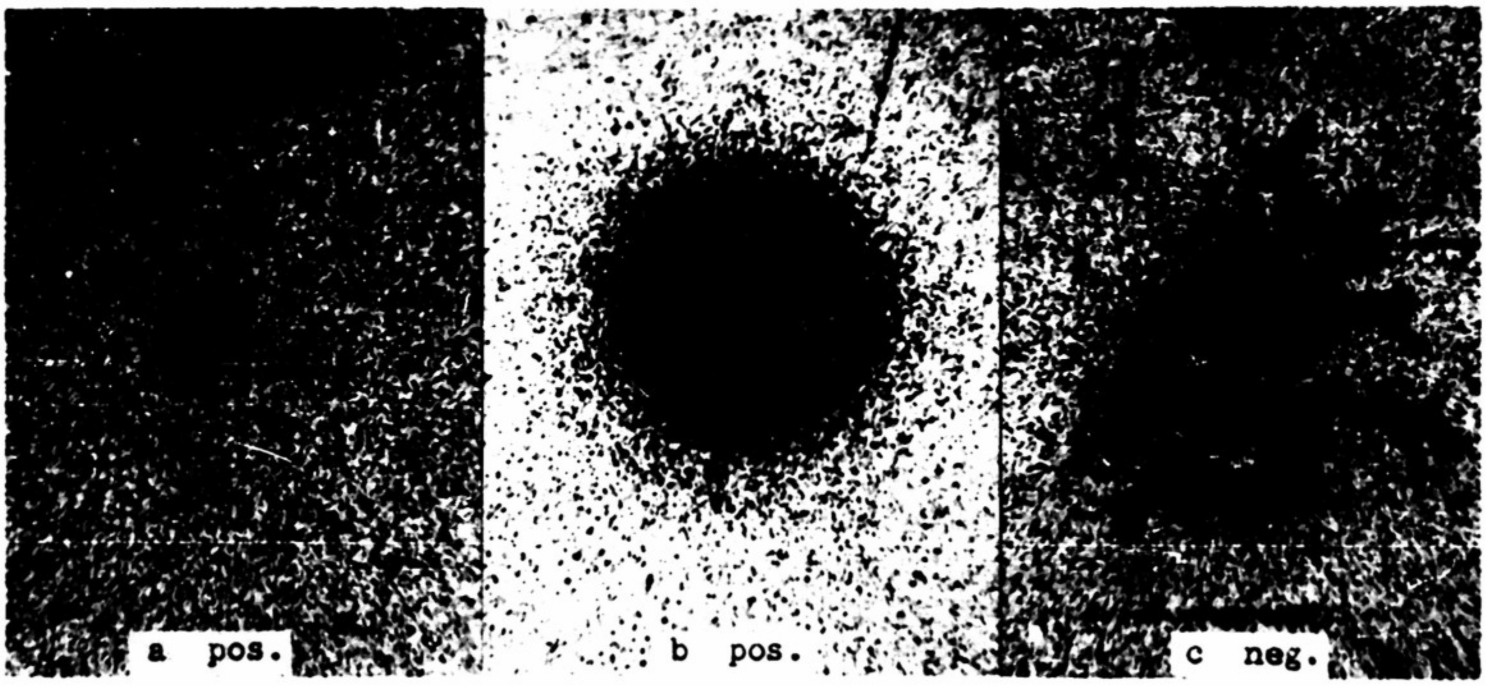
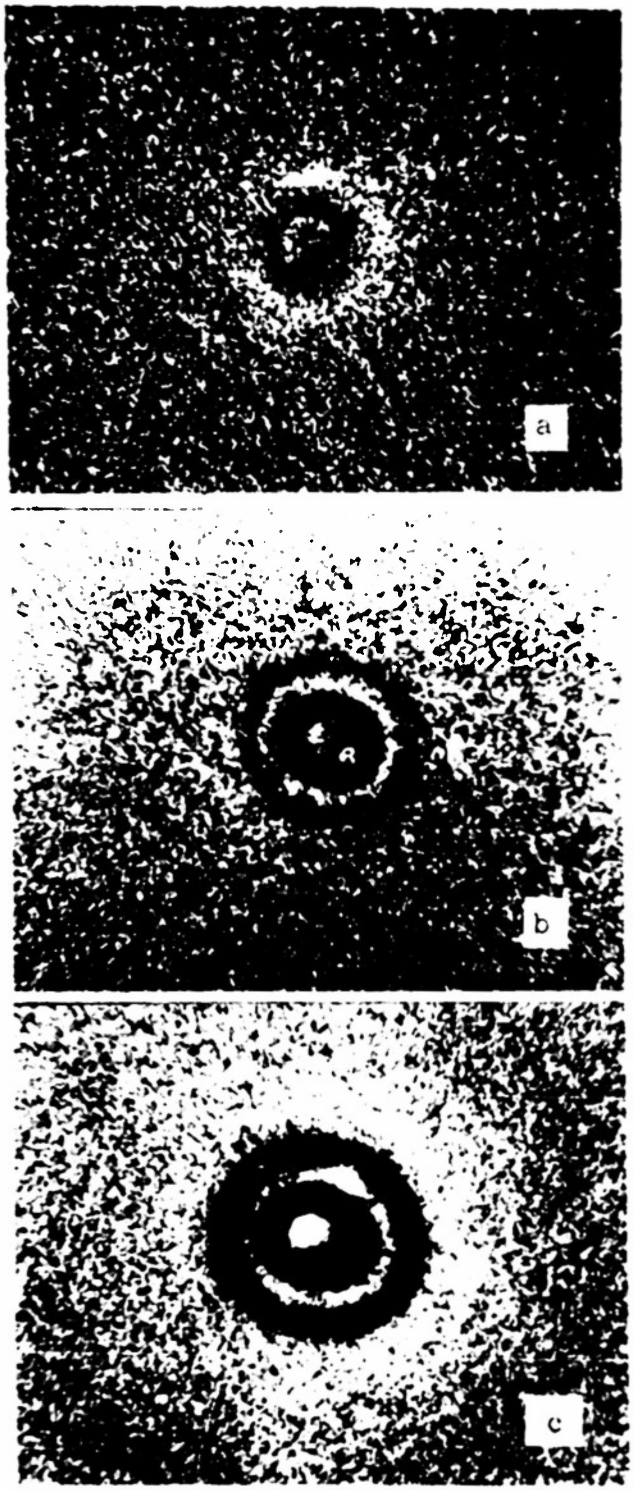


Fig. 48 Needle electrodes in contact with emulsion

100X



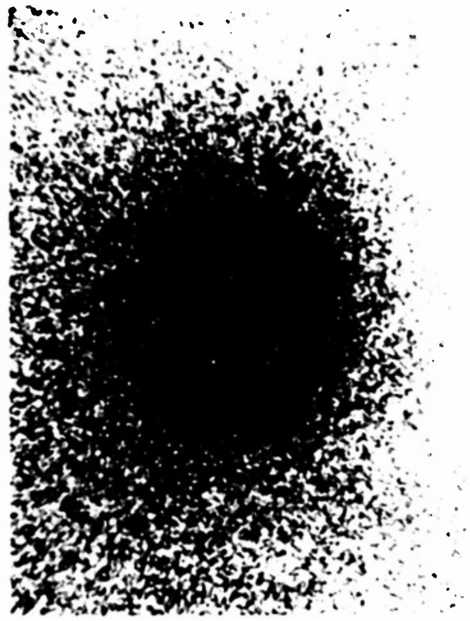


Fig. 50 Geiger discharges taken under conditions similar to the ones in Fig. 49

Fig. 49 Electrical effect underneath anode

REFERENCES

1 Sturm, R. E. and Morgan, R. H., Am. J. Roentgenology, 62 (1949) Nov.

2 O'Connor, D. T. and Polansky, D., Non-destructive Testing, Fall 1951, p. 10

3 Coltman, J. W., Radiology, 51, (1948), 359

4 Westinghouse X-ray Division, Rev. Scient. Instr., 19, (1948), 477, #7

5 Lusby, W. S., Electr. Engng., 70, (1951), 292

6 Lusby, W. S., Non-destructive Testing, Winter 1952, p. 7

7 Drosd, R. D., Liddiard, T. P., Singleton, B. N. Jr., Navord Rep., 1811, (1951), Nov. 23

8 Gibson, F. C., Abstr. 5th Conference of Basic Instrumentation, N.B.S., 1952, p. 23

9 Moon, R. J., Am. J. Roentgenology, 59, (1948), 886, #6

10 Hodges, P. C., and Moon, R. J., Am. J. Roentgenology, 62, (1949), 637, #5

11 Moon, R. J., Science, 112, (1950), 389, #2910

12 Hodges, P. C., and Skaggs, L. S., Am. J. Roentgenology, 66, (1951), 705

13 Morgan, R. H., Am. J. Roentgenology, 62, (1949), 644

14 Mayneord, W. V. and Belcher, E. H., Brit. J. Radiology, 1950, 259, #22

15 Mayneord, W. V., Turner, R. C., Newbery, S. P., and Hodt, M. J., Nature, 168, (1951), 762

16 Haworth, F. E., U. S. Patent 2,457,555, Dec. 1948

17 Selényi, P., Zs. f. Physik, 47, (1928), 895

18 Carlson, C. F., U. S. Pat. 2,221,776; 2,277,013; 2,297,691

19 Langer, N., Radio News, Radio-Electronic Engng., 3, (1944), 22

20 McMaster, R. C. and Schaffert, R. M., Non-destructive Testing, Summer 1950, p. 11

21 McMaster, R. C., Non-destructive Testing, 10, (1951), 8, #1

22 Goldmann, R., Brit. Patent 464,112 (1935)

23 Greinacher, H., Helv. Physica Acta, 20, (1947), 222

24 Johnson, T. H., Phys. Rev., 75, (1949), 1301

25 Merrill, F. H. and Von Hippel, A., J. Appl. Physics, 10, (1939), 873

26 Keinath, G., Arch. Techn. Messen, J 835-1 (1932)

27 Gemant, A., Zeitschr. Techn. Physik, 12, (1931), 250

28 Gemant, A., World Power, 24, (1935), 8

29 Thomas, A. M., World Power, 24, (1935), 87

30 Akahira, T. and Kamazawa, M., Sc. Pap. Inst. Phys. Chem. Res. Tokyo, 36, 910, 1939

31 Mason, J. H., Nature, 164, (1949), 451

32 Prat. S., and Schlemmer, J., Sbornik A. P., 14-80, (1940), 1

33 Zahradnicek, J., Publ. Fac. Sc. Univ. Masaryk, 150, (1948), 3

34 Destriau, G., Phil. Mag., 38, (1947), 700, 774, 880

35 Payne, E. C., Mager, E. L., and Jerome, C. W., Ilum. Engng., 45, (1950), 668

36 Waymuth, J. F., Jerome, C. W., and Gungle, W. C. Sylvania Technol., 3, (1952), 54

37 Payne, E. C., Mager, E. L., and Jerome, C. W., Sylvania Technol., 4, (1951), 2

38 Klemperer, O., Einfuehrung i. d. Elektronik, Springer, Berlin, 1933, p. 37

39 Becker, A. and Kipphan, E., Ann. d. Physik, 10, (1931), 15

40 Ellis, C. D., and Wooster, G. H., Proc. Roy. Soc., 114, (1927), 266

41 Silberstein, L., Phil. Mag., 45, (1923), 1062

42 Klemperer, O., Einfuehrung i. d. Elektronik, Springer, Berlin, 1933, p. 41

43 Bainbridge, K. T., J. Franklin Inst., 212, (1931), 489

44 Garlick, G., Luminescent Materials, Oxford, 1949

45 Friedman, H., Proc. I.R.E., 37, (1949), 791

46 Brown, S. C., Phys. Rev., 62, (1942), 244

47 Brown, S. C., and Miller, W. W., Rev. Sci. Inst., 18, (1947), 496

48 Raether, H., Ergebnisse d. exakt. Naturw., 22, (1949), 73

49 Liesegang, R. E., Naturw. Wochenschr., 11, (1396), 353